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Full length article

## The variability of mass concentrations and source apportionment analysis of equivalent black carbon across urban Europe

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## ABSTRACT

This study analyzed the variability of equivalent black carbon (eBC) mass concentrations and their sources in urban Europe to provide insights into the use of eBC as an advanced air quality (AQ) parameter for AQ standards. This study compiled eBC mass concentration datasets covering the period between 2006 and 2022 from 50 measurement stations, including 23 urban background (UB), 18 traffic (TR), 7 suburban (SUB), and 2 regional background (RB) sites. The results highlighted the need for the harmonization of eBC measurements to allow for direct comparisons between eBC mass concentrations measured across urban Europe. The eBC mass concentrations exhibited a decreasing trend as follows: TR > UB > SUB > RB. Furthermore, a clear decreasing trend in eBC concentrations was observed in the UB sites moving from Southern to Northern Europe. The eBC mass concentrations exhibited significant spatiotemporal heterogeneity, including marked differences in eBC mass concentration and variable contributions of pollution sources to bulk eBC between different cities. Seasonal patterns in eBC concentrations were also evident, with higher winter concentrations observed in a large proportion of cities, especially at UB and SUB sites. The contribution of eBC from fossil fuel combustion, mostly traffic (eBC<sub>T</sub>) was higher than that of residential and commercial sources (eBC<sub>RC</sub>) in all European sites studied. Nevertheless, eBC<sub>RC</sub> still had a substantial contribution to total eBC mass concentrations at a majority of the sites. eBC trend analysis revealed decreasing trends for eBC<sub>T</sub> over the last decade, while eBC<sub>RC</sub> remained relatively constant or even increased slightly in some cities.

## 1. Introduction

Black carbon (BC) is a primary particulate pollutant generated by combustion processes (Bond et al., 2013); it is one of the key components of ambient particulate matter (PM). BC emissions originate from several anthropogenic combustion sources, such as the diesel engines of on-road and off-road vehicles (i.e., transportation and industry), maritime shipping (Geels et al., 2021; Lack and Corbett, 2012; Zhang et al., 2019a; Zhang et al., 2019b), domestic solid fuel (biomass-burning and coal combustion), and other combustion processes (e.g., residential heating) (Bond et al., 2013; Briggs and Long, 2016; Intergovernmental Panel on Climate Change [IPCC], 2021).

BC terminology is diverse and there are many uncertainties associated with the definitions and methods used to measure BC (Petzold et al., 2013; Grange et al., 2020). Several previous studies have proposed varying definitions and recommended standard terminology for carbonaceous aerosols, and the nomenclature used for BC can be confusing (Bond et al., 2013; Petzold et al., 2013). Equivalent BC (eBC) is commonly defined as the mass concentration of BC as indirectly determined by light absorption techniques such as filter-based absorption photometers (Petzold et al., 2013). Filter-based absorption photometers measure the attenuation ( $\sigma_{ATN}$ ) or transmission of light through a particle-loaded filter tape and convert them to the absorption of light. These optical instruments, such as the aethalometer (AE) and the multi-angle absorption photometer (MAAP), have been widely used to estimate eBC mass concentrations from the amount of light absorbed by the particles collected on filter tape samples using a conversion factor known as a mass absorption cross-section (MAC). These techniques thus indirectly determine the eBC mass concentration based on particle light absorption coefficient measurements at specific wavelengths. Thus, eBC is defined operationally by the method used for its measurements (Moosmüller et al., 2009; Petzold et al., 2013). Consequently, the term eBC will be used throughout this study as all eBC data were calculated using filter-absorption photometers and their respective instrument software.

Unlike most components of atmospheric aerosol that have a climate cooling effect, eBC exhibits a substantial positive radiative forcing (warming) effect due to its strong light-absorption, albeit with significant quantitative uncertainties (Bond et al., 2013; IPCC, 2021; Ivančić et al., 2022; Lack and Cappa, 2010). There are no generally agreed standards on eBC measurement methods and their related uncertainties (Petzold et al., 2013). Previous intercomparisons of aerosol absorption

photometers have shown that there is a significant variation in the light-absorption response of aerosol particles on different instruments due to source contributions, sample flow, and spot size (Cuesta-Mosquera et al., 2021; Müller et al., 2011). The Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) (Measurement guidelines, 2022) and Working Group WG 35 of the European Committee for Standardization (CEN) have attempted to provide recommendations in order to harmonize eBC measurements. However, the need for a broad implementation of these recommendations is evident from the wide array of protocols used to measure eBC across Europe.

eBC-related research in urban areas attracted a significant amount of attention due to the potential health impacts of soot (i.e., combustion particles made of a core of eBC and coating of organic and sulfur compounds) of which eBC is a tracer (Kumar et al., 2020; Segersson et al., 2017). The adverse impacts of eBC on public health and climate—at both local and global scales—have been extensively documented (Briggs and Long, 2016; World Health Organization [WHO], 2021). Nevertheless, available data on eBC and associated health effects have not been sufficient to provide consistent results to set up or discard air quality (AQ) guidelines (WHO, 2021). Furthermore, eBC itself does not necessarily have a negative impact on public health; rather, it is the presence of other species condensed onto eBC particles that may cause health issues (WHO, 2012). Nevertheless, it has been concluded that eBC is a better indicator of harmful particulate substances from combustion sources (especially traffic) than the mass of undifferentiated atmospheric PM. Thus, the closer monitoring of eBC may improve the assessment of policy actions to abate air pollution from combustion sources while also providing more consistent data for epidemiological analysis (Geng et al., 2013; WHO, 2012, 2021).

Several studies have investigated the variability of eBC mass concentrations in specific urban, traffic, and regional background environments. Some of these also focused on the source apportionment (SA) of eBC, such as the proportion of eBC that stems from the burning of fossil fuels in vehicular exhaust emissions (road traffic; eBC<sub>T</sub>) and solid fuel combustion (mainly from residential and commercial wood and coal burning; eBC<sub>RC</sub>) (Blanco-Donado et al., 2022; Grange et al., 2020; Helin et al., 2018; Liakakou et al., 2020; Mbengue et al., 2020; Milinković et al., 2021; Minderytė et al., 2022; Mousavi et al., 2018, 2019, Sandradewi et al., 2008a,b). These studies have been individually reported on the climatology, characterization, and variability of eBC in different regions (Crilley et al., 2015; Favez et al., 2009; Grange et al., 2020; Helin et al., 2018; Jafar and Harrison, 2021; Krecl et al., 2011;

Kutzner et al., 2018; Laborde et al., 2013; Luoma et al., 2021; Lyamani et al., 2011; Mousavi et al., 2019; Pandolfi et al., 2014; Schaap and Denier van der Gon, 2007; Singh et al., 2018; Titos et al., 2017; Vanderstraeten et al., 2011). However, to the best of our knowledge, a large-scale database that allows for access to eBC data across urban Europe has not been implemented.

The Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in EU Urban & Industrial AreaS (RI-URBANS) project is a European H2020-Green Deal initiative that aims to provide tools for the measurement and analysis of advanced AQ parameters (RI-URBANS, 2020), including eBC, ultrafine particles, and oxidative potential in urban environments, which will consequently allow for the enhanced assessment of AQ policies. Thus, it is necessary to harmonize measurement, instrumentation, and data analysis protocols to provide comparable data from different cities across Europe.

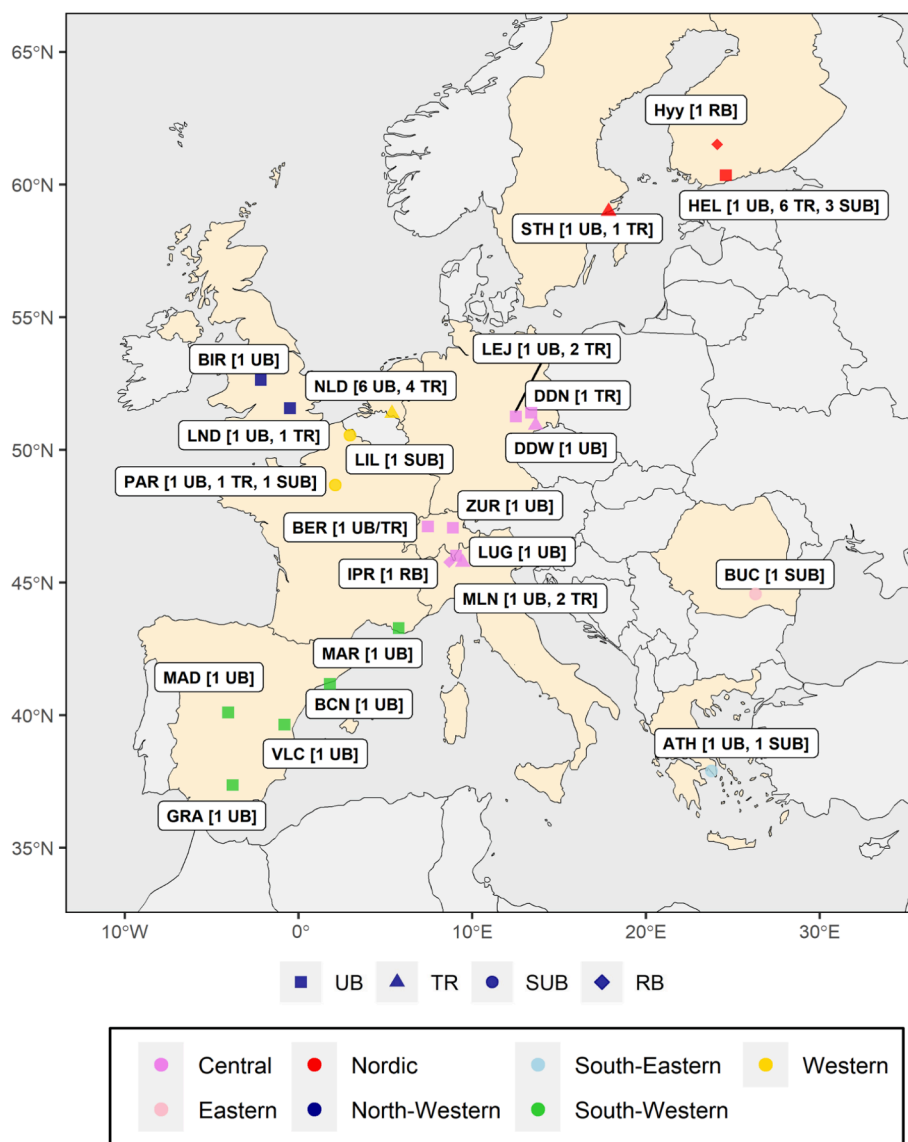
One of the objectives of RI-URBANS is to evaluate the application of advanced AQ metrics, such as eBC, in urban environments and other air pollution hotspots. In line with this objective, this study aims to 1) compile and interpret long-term ambient eBC measurements across urban Europe; 2) analyze the current measurement corrections, concentration levels, and spatial and temporal variability covering a wide

variety of climate and urban patterns; and 3) quantify the contribution of  $eBC_T$  and  $eBC_{RC}$  to bulk urban eBC mass concentrations. This long-term eBC observations provide valuable information that can be used to evaluate its potential to improve policy assessments as an AQ standard as well as better protect human health (Irwin et al., 2015; Janssen et al., 2011; WHO, 2012). The harmonized urban eBC data compiled in this study will be used in epidemiological evaluations, for model validation, and for improving emission inventories in future investigations.

## 2. Methodology

### 2.1. Measurement sites

This study aimed to evaluate the long-term phenomenology of eBC by compiling datasets on eBC mass concentrations from 50 European monitoring sites, covering various periods between 2006 and 2022. These 50 measurement sites include 23 urban background (UB) sites, 18 traffic (TR) sites, 7 sub-urban background (SUB) sites, and 2 regional background (RB) sites across 29 cities in 11 European countries. These categories were selected based on the descriptions used for the classification of AQ monitoring sites (European Environmental Agency [EEA],



**Fig. 1.** The location and type of monitoring sites that have supplied eBC data to this study. UB: urban background; SUB: suburban background; TR: traffic; RB: regional background. The colors correspond to the seven main geographic regions around Europe.

2023; UK AIR, 2023). As this study focuses on European urban environments, a few suburban and regional background sites were selected to ensure better spatial coverage and a greater understanding of eBC variability. However, it should be noted that their eBC trends cannot be considered to be representative of the variability for all European RB sites, as previously reported by Laj et al. (2020) or Collaud Coen et al. (2020). Fig. 1 describes the distribution of monitoring sites that are currently providing measurements of eBC mass concentrations. The measurement sites were grouped into seven categories based on their geographical location (Northern, Eastern, Western, Central, South-Eastern, South-Western, and North-Western); this categorization scheme follows from the grouping of sites in previous European phenomenology studies (Pandolfi et al., 2018; Putaud et al., 2010). The highlighted areas in Fig. 1 are indicative of geographical classifications.

The data coverage mainly comprises UB and TR sites, located in cities from different climate zones of Europe that are affected by different road traffic volumes, industrial sources, power plant emissions, harbors, domestic sources, etc. As shown in Fig. 1, the spatial coverage over Europe is not homogeneous: there is a higher density of measurement sites over central Europe and a much lower density of sites in Eastern Europe—this is denoted by the grey area in which eBC measurements were not available.

## 2.2. Instrumentation

Table S1 summarizes the instruments used to measure eBC mass concentrations in the 50 sites. In addition to differences in instrumentation, relevant distinct measurement settings have also been noted (e.g., different sampling inlet heights). eBC mass concentrations were primarily monitored by filter-based absorption photometers. eBC data were obtained through the use of MAAPs (Thermo Scientific, discontinued) or aethalometers (mainly AE33 model; Magee Scientific). The recommended terminology and measurement techniques for these instruments have been comprehensively discussed in Petzold and Schönlinner (2004), Petzold et al. (2005), and Drinovec et al. (2015).

The MAAP was developed to decrease the uncertainties in absorption measurements ( $b_{\text{abs}}$ ). It measures the transmitted, backscattered light through the aerosol-loaded filter tape and then applies a radiative transfer scheme to convert the measurements into particle light absorption coefficients. It determines the particle light absorption coefficient using a 670 nm operating wavelength (as specified by the manufacturer), while its actual wavelength is 637 nm (Müller et al., 2011). The default MAC setting of  $6.6 \text{ m}^2 \text{ g}^{-1}$  and a nominal wavelength of 670 nm are used to convert the optically measured absorption data to eBC mass concentrations. The instrument also accounts for filter-loading-related artifacts that can affect the calculation of the absorption coefficient (Petzold and Schönlinner, 2004). However, given the discontinued production of the MAAP, its future availability for long-term observations is uncertain. Furthermore, MAAP data were not accessible from all 50 monitoring sites investigated in this study, as some sites deployed other types of absorption photometers, as discussed in the following section.

The AE33 dual-spot Aethalometer (Magee Scientific) measures the attenuation of light ( $\sigma_{\text{ATN}}$ ) through an aerosol-loaded filter tape samples and provides particle light absorption coefficients at seven different wavelengths (370, 470, 520, 590, 660, 880, and 950 nm). It converts the optical absorbance into eBC mass concentrations using a default MAC value of  $7.77 \text{ m}^2 \text{ g}^{-1}$  at 880 nm. AE33 has been equipped with a real-time filter loading effect compensation algorithm based on the dual-spot measurements (Drinovec et al., 2015). Older Aethalometer models such as the AE31 and AE22 measure light attenuation through the filter at seven (AE31) and two (AE22; 370 and 880 nm) wavelengths using an instrument-derived mass attenuation cross section value of  $16.62 \text{ m}^2 \text{ g}^{-1}$  at 880 nm. However, these models require correction for loading effects, and algorithms for correcting this artifact have been developed and tested by different researchers (Collaud Coen et al., 2020;

Virkkula, 2021; Virkkula et al., 2007; Weingartner et al., 2003).

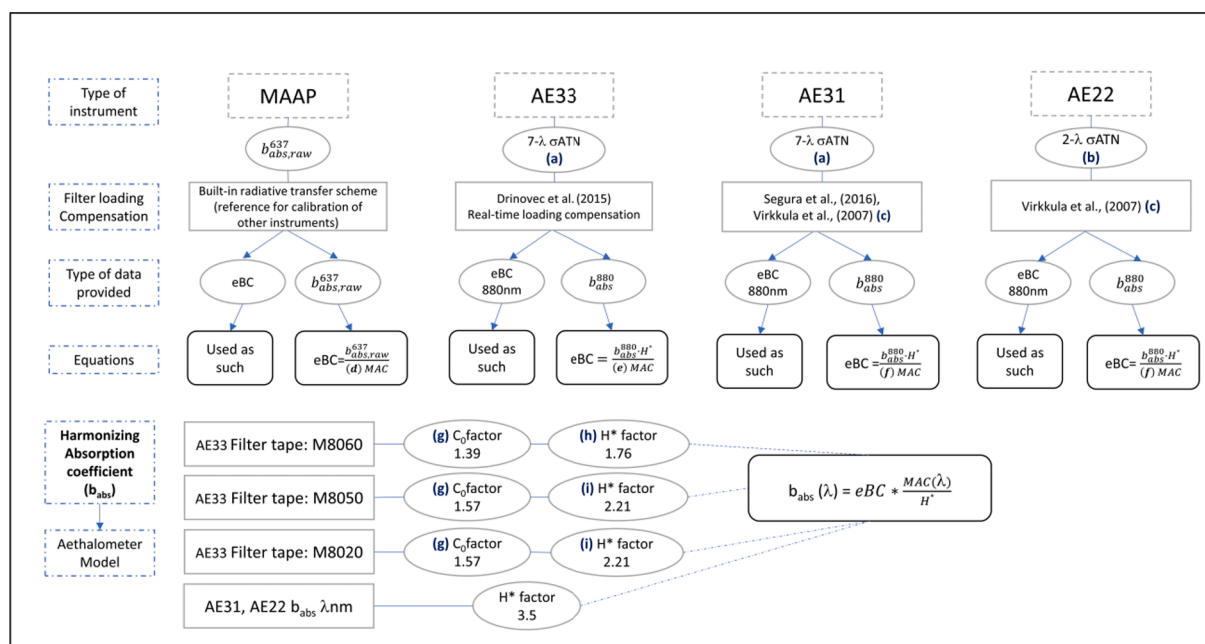
In brief, of the 50 observation sites present across Europe, the MAAP was the most commonly used instrument for measuring eBC mass concentrations, used at 30 of the 50 sites. The MAAP was primarily used in measurement sites across Western and Northern Europe (see Table S1). Both a MAAP and an aethalometer were deployed in parallel at a few sites (BCN, GRA, STH, MLN, and SMR). In the RI-URBANS database, data from 23 sites were obtained using the AE33 instrument, while only a few stations provided data from AE22 and AE31 instruments (e.g., LND, SMR, and VLC). Table S1 also provides information about the geographic region, site, city, country, station type, coordinates, altitude, measurement height, data coverage, and type of data received (more details can be found in Section 2.3).

## 2.3. Data treatment and pre-processing

This study required the use of station metadata, including station information (e.g., type, altitude, height, and coordinates), the filter absorption photometer used, the measurement method, and any relevant operational settings (e.g., filter tape, inlet, flow rate, and reference MAC). Although each monitoring site was active for different periods, this study primarily focused on the period between 2017 and 2019 to evaluate the spatial variability of eBC mass concentration to 1) ensure that data was available from all sites and 2) to avoid the effects of the COVID-19 lockdowns. However, only sites that provided more than nine years of eBC observations and where at least 75% of the data was available were considered for trend analysis; as a result, the entire dataset can be used for subsequent epidemiological studies. It should be noted that missing data in the datasets were not gap-filled in further analyses. Various statistical measures were calculated for each site, including the minimum, maximum, 25th, 75th, and 99th percentiles, mean, median, and the number of data points per station. The overall dataset covers the period between 2006 and 2022, with single-site coverage ranging from a year to 14 years. Fig. S1 visualizes the distribution of eBC data available in the RI-URBANS dataset, including data coverage, data capture, and missing data from the various instruments. The blank area represents the missing data across the entire period. Aside from the low data coverage of some stations, the common period used in this study for the analysis and interpretation of the results (2017–2019), is fulfilled by majority of the sites.

The majority of the collected data were directly supplied by the data providers as hourly averages (Table S1). There are only two cases in which raw data with a native time resolution were provided; these data were averaged hourly for the purposes of this study. In some cases, EBAS Level 2 quality assured/quality checked (QA/QC) data were directly downloaded from the EBAS database (EBAS, <https://ebas-data.nilu.no/>). The data were processed following the ACTRIS recommendations for the reporting of eBC measurements (Müller and Fiebig, 2018a, 2018b), ensuring the comparability of eBC measurements across the sites by employing harmonized measurement protocols.

The procedure used to obtain particle light absorption coefficients—and consequently, eBC concentrations—from the available data is summarized in Fig. 2. The majority of the sites provided eBC concentrations that were used as is to analyze eBC variability. In cases where particle light absorption data were provided, the eBC concentrations were retrieved using default MAC values and the opportune harmonization factors  $H^*$  as described in the following. Fig. 2 also presents the equations that were used to calculate the particle light absorption coefficients from the wavelength-dependent eBC concentrations provided by aethalometer instruments. These harmonized particle light absorption coefficients were then used in the aethalometer model for eBC source apportionment. As previously mentioned, AE33 instruments already correct online for factor loading, while data from older aethalometers (AE31 and AE22) were corrected offline by data providers. Specifically, the correction scheme proposed by Segura et al. (2016) was applied to the AE31 data from VLC\_UB, while the scheme



**Fig. 2.** The overall scheme by which to process corrected and harmonized eBC mass concentrations and absorption coefficients ( $b_{abs}$ ) from the attenuation coefficient ( $\sigma_{ATN}$ ). Data were supplied from different types of instruments and methods, which were then used in eBC source apportionment using the harmonized absorption coefficient ( $b_{abs}$ ). More details are provided in section 2.3. (a) AE33 and AE31 wavelengths: 370, 470, 520, 590, 660, 880, and 950 nm; (b) AE22 wavelengths: 370 and 880 nm; (c) Offline filter loading correction algorithms: Segura et al. (2016) was used to correct the AE31 data from Valencia; Virkkula et al. (2007) was used to correct AE31 data from SMR\_RB and correct AE22 data from LND\_UB and LND\_TR; (d) MAAP: MAC = 6.6 m<sup>2</sup>/g at 670 nm; (e) AE33: MAC = 7.77 m<sup>2</sup>/g at 880 nm; (f) AE31 and AE22: mass attenuation cross section = 16.62 m<sup>2</sup>/g at 880 nm; (g) Default multiple-scattering enhancement correction factor used by the aethalometer software (1.39 or 1.57); (h) Harmonization factor ( $H^*$ ) for M8060 filter tape; (i) Harmonization factor ( $H^*$ ) for M8050 and M8020 filter tapes. M8050 was used for a short time and was not characterized; thus, processing was performed with the same harmonization factor as filter tape M8020.

proposed by Virkkula et al. (2007) was applied to the AE31 and AE22 data from SMR\_RB and LND\_UB/LND\_TR, respectively (Fig. 2).

To calculate the particle absorption coefficients from aethalometer instruments, two parameters that depend on the type of aethalometer and filter tape used must be considered: the default multiple-scattering enhancement parameter ( $C_0$ ) and the standardized harmonization factor ( $H^*$ ) provided by ACTRIS (Müller and Fiebig, 2018a). For AE33 instruments, the  $C_0$  is manually set using the instrument software and should be changed depending on the filter tape used. Three different filter tapes were used across the monitoring sites: the M8020 (also known as TFE-coated fiberglass tape or T60A20), which was employed from approximately 2011 until February 2016; the M8050, which was used between February 2016 and October 2017; and the recently recommended M8060 filter tape, which has been in use since October 2017. The default  $C_0$  values are 1.57 for both M8020 and M8050 and 1.39 for the new M8060 filter tape. However, recent studies have shown that these default  $C_0$  values were not appropriate; thus,  $H^*$  factors were introduced to standardize the calculation of the absorption coefficients. ACTRIS proposed an  $H^*$  value of 1.76 for M8060 filter tape, while the proposed  $H^*$  value for the AE33 M8020 and M8050 filter tapes was 2.21 (Müller and Fiebig, 2018a). Currently, guidelines from the Global Atmosphere Watch Programme suggest the use of  $H^* = 3.5$  for AE31 and AE22 (WMO/GAW, 2016). Thus, it is crucial to have the information about the filter tape used as well as the default  $C_0$  to apply these corrections appropriately.

It should be considered that  $H^*$  can vary depending on the physical properties of particles collected on the filter tape. For example, Yus-Díez et al. (2021) showed that the  $C_0$ , and consequently the  $H^*$ , increases when the single scattering albedo (SSA) of collected particles is high (>0.95). Yus-Díez et al. (2021) observed this effect, known as cross-sensitivity to scattering, at two remote/regional background sites but did not observe it at the urban site, where the SSA was low. Thus, the assumption of a constant  $H^*$  is reasonably correct at the urban/traffic

sites included in this work.

As previously mentioned, we have presented the eBC concentrations as calculated according to the default MAC values provided by the manufacturers as this is the common practice in monitoring networks (cf. Fig. 2). However, it should be acknowledged that numerous studies have shown that the MAC can vary between different sites as well as over time depending on the physicochemical properties of the eBC particles (Cho et al., 2019; Ciupek et al., 2021; Grange et al., 2020; Knox et al., 2009; Kondo et al., 2009; Mbengue et al., 2021; Srivastava et al., 2022; Wu et al., 2021; Yuan et al., 2021; Zanatta et al., 2016; Zhao et al., 2021).

Thus, it should be noted that the comparability of eBC data reported in this study is partially affected by instrument-to-instrument variability. For example, Müller et al. (2011) reported a unit-to-unit variability of <5% for MAAP instruments while noting a variability of up to 30% for old aethalometer models, such as the AE31. This large variation was primarily associated with sample flow, spot size, and the inadequacy of current correction functions. As previously mentioned, AE31 data needs to be corrected off-line for factor loading effects, thus contributing to the higher noise inherent in AE31 data compared to MAAP data. For example, Asmi et al. (2021) reported a difference of about 7% between MAAP and AE31 data with a low correlation ( $R^2 = 0.65$ ); in contrast, the correlation between MAAP and AE33 instruments was much higher ( $R^2 = 0.87$ ). Additionally, Helin et al. (2018) reported a good correlation ( $R^2 > 0.90$ ) between MAAP and AE33 data, with AE33 instruments reporting slightly higher concentrations. Recently, Cuesta-Mosquera et al. (2021) reported on the relatively small unit-to-unit variability of AE33-based eBC measurements, ranging between -16% and 8% with an average deviation of around -1% for soot. In most cases, the deviations could be explained by the type of filter material employed by the instruments, the total particle load on the filter, and the flow calibration. To reduce the instrument-to-instrument variability of AE33, Cuesta-Mosquera et al. (2021) recommended the use of the most

recent version of filter tape (M8060) while avoiding the use of older filter tapes. Furthermore, they highlighted the importance of reporting not only the filter tape used but also the corresponding multiple-scattering parameter  $C_0$  set by the internal aethalometer settings. Due to its high accuracy and low uncertainty (12%; [Petzold et al., 2005](#)), the MAAP is considered to be the ideal reference instrument for the calculation of particle light absorption coefficients. Thus, we used eBC data from the MAAP instruments in stations where other filter absorption photometers were simultaneously deployed (IPR\_RB, SMR\_RB, BCN\_UB, and GRA\_UB).

Long-term trends were studied using three different approaches. The first approach involves the use of a Theil-Sen slope regression estimator: a robust, reliable, and non-parametric estimator provided in the Openair R package ([Carslaw and Ropkins, 2012](#)). This method assesses long-term trends by calculating monthly means as derived from hourly measurements. This approach not only enables the determination of statistical significance but also facilitates the estimation of slope values for the identified trends. As previously mentioned, only monitoring sites with a minimum of nine years of eBC mass concentration data with at least 75% valid measurements over that period were used in these analyses. It is important to note that some sites experienced changes in the measurement methodology over the nine-year period, most commonly due to changes in the filter-based instruments being deployed (e.g., shifting from MAAP to AE33 or from AE31 to AE33). Thus, the trends in absorption measurements can be influenced by the change in signals due to different instruments in 3 out of 13 sites (e.g., ZUR\_UB, BER\_UB, and SIR\_SUB). Theil-Sen method was primarily used for inter-annual trend analysis. However, it does not take the shape of trends and possible breakpoints into account. Thus, the shape of trends and their seasonal variations were investigated through the more widely used decomposition method, a seasonal-trend decomposition time series procedure (STL) based on the 'Loess' estimator ([Cleveland et al., 1990](#)). The STL method is a powerful and versatile approach for analyzing time series data, allowing for the extraction of trends and seasonal components while accounting for various data characteristics and patterns. 95% confidence intervals calculated using bootstraps are also provided for the STL and Theil-Sen results. Another approach to investigate the presence of potential breakpoints that split the trends into segments with different slopes was piecewise regression using the "segmented" package ([Muggeo, 2003, 2008](#)). To estimate the slope and statistical significance of each segment between breakpoint positions, an interactive procedure that involves approximate starting values for the breakpoints and a bootstrap restarting methodology was implemented to enhance the robustness of the analysis and obtain standard errors ([Wood, 2001](#)). By using this procedure to fit a piecewise regression model, we were able to effectively characterize and interpret the distinct trends observed in the eBC data. This approach provided valuable insights into the changes and shifts in eBC mass concentration, allowing for a comprehensive analysis of the behavior of the data over different segments.

#### 2.4. Aethalometer model and source apportionment

To evaluate the relative contributors of eBC<sub>T</sub> and eBC<sub>RC</sub> at the RI-URBANS sites, an aethalometer model was employed; this uses the multi-wavelength absorption data obtained from the AE33 following a procedure initially developed by [Sandradewi et al. \(2008a\)](#). Briefly, the model uses a bilinear method to estimate both eBC sub-fractions. This model has been widely applied and used by numerous investigators, discriminating between mostly traffic (notably including diesel, gasoline and natural gas combustion) and residential and commercial (biomass or coal combustion) eBC components ([Becerril-Valle et al., 2017](#); [Favez et al., 2010](#); [Grange et al., 2020](#); [Harrison et al., 2013](#); [Helin et al., 2018](#); [Tobler et al., 2021](#); [Virkkula, 2021](#)). The AE model was applied using 23 AE33 datasets taken from the RI-URBANS database. The source apportionment analysis used time series data between 2017 and 2019. The aethalometer model uses pre-defined source-specific absorption

Ångström exponents (AAEs) as recommended by [Sandradewi et al. \(2008a\)](#) and [Zotter et al. \(2017\)](#).

The AAE is an optical parameter that describes the spectral dependence of light absorption by aerosols. It has been intensively employed for eBC source apportionment and aerosol characterization (e.g., BC, Brown Carbon "BrC", and dust) ([Garg et al., 2016](#); [Li et al., 2016](#); [Liu et al., 2018](#); [Wang et al., 2021](#); [Zhang et al., 2019a](#); [Zhang et al., 2019b](#)). An AAE of around 1 indicates that the absorption is dominated by BC; a progressive increase in AAE is associated with an increasing contribution from non-BC absorbing components such as BrC or mineral dust ([Zhang et al., 2020a, 2020b](#)). The application of the aethalometer model requires a priori knowledge of the AAE of the main sources of BC. Values of around 1 and 2 for the two wavelengths of 470 and 950 nm are commonly used for traffic and residential, commercial sources (e.g., biomass burning and coal combustion), respectively ([Sandradewi et al., 2008a](#)). More recent estimates based on <sup>14</sup>C analysis have suggested values of 0.90 and 1.68 ([Zotter et al., 2017](#)). However, both AAE values may vary significantly depending on the type of fuels being burnt, burning conditions, and the microphysical properties of the BC particles (the size of the BC particle and internal mixing with non-absorbing or less absorbing material): a single value cannot be representative of the whole measurement period considered ([Helin et al., 2021](#); [Li et al., 2016](#); [Liu et al., 2018](#)). For this reason, the aethalometer source apportionment model is associated with uncertainties related to fixed specific AAEs of traffic-related and residential and commercial emission sources [AAE<sub>T</sub> and AAE<sub>RC</sub>] ([Harrison et al., 2013](#); [Helin et al., 2021](#); [Zotter et al., 2017](#)).

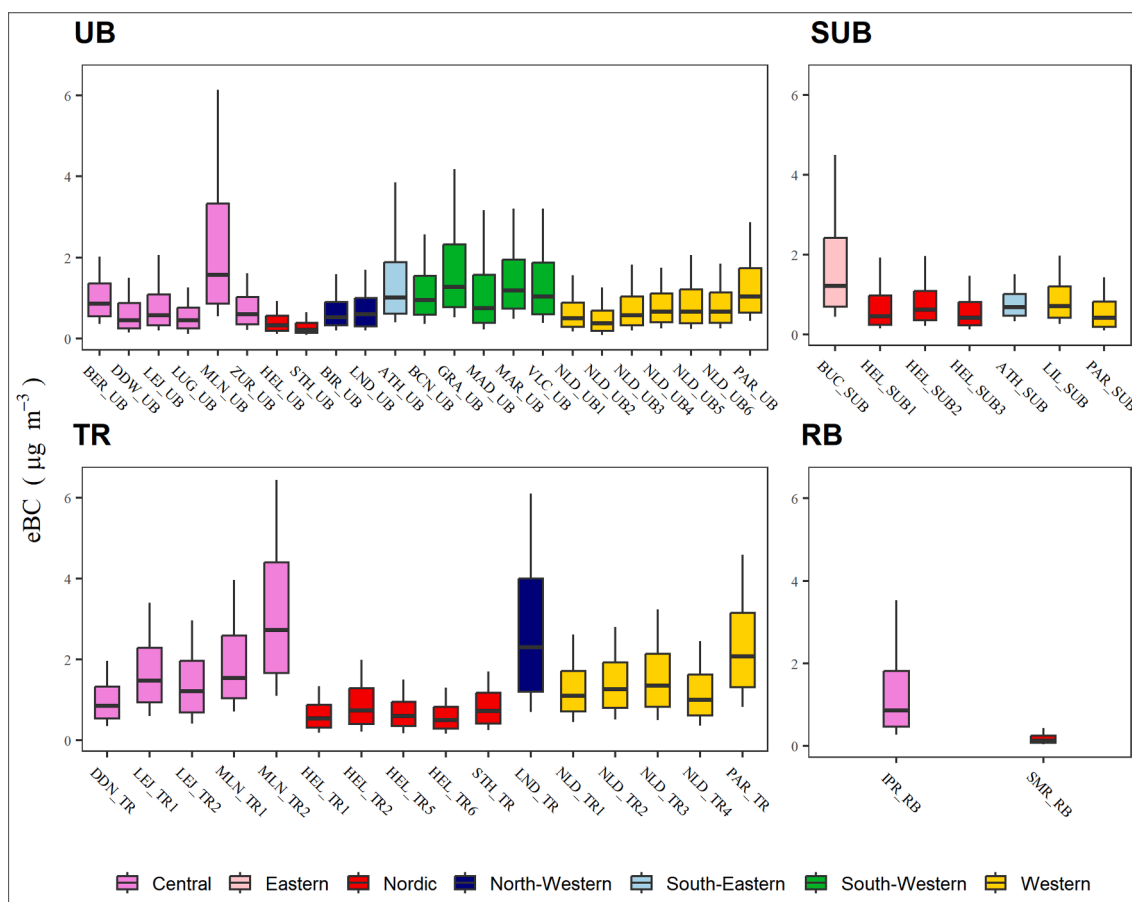
Thus, the selection of robust AAE values is a critical component of the aethalometer model and has been validated in previous studies using <sup>14</sup>C observations as well as other auxiliary measurements such as Elemental Carbon, Organic Carbon (EC/OC) ([Sandradewi et al., 2008a](#); [Sandradewi et al., 2008b](#); [Zotter et al., 2017](#)). This study follows the conventional approach of using fixed AAE values for the source apportionment of AE33 observations due to the lack of ancillary measurements (such as <sup>14</sup>C measurement data) that could be used to determine site-dependent AAEs. Thus, AAE values of 1 and 0.90 were used for eBC<sub>T</sub>, while AAE values of 2 and 1.68 were used for eBC<sub>RC</sub> for the wavelengths of 470 nm and 950 nm, respectively. The results obtained were compared to evaluate the variations across different regions due to the limitation of constant AAE values.

### 3. Results and discussion

#### 3.1. Overview of eBC observations and average eBC mass concentrations

This section reports the measurement data collected using different filter-based techniques at 50 European sites. [Table S2](#) presents the statistical analysis of the eBC mass concentration at each site using hourly averaged data recorded between 2017 and 2019. Specifically, the mean, maximum, minimum, and standard deviation were calculated for each site. Data coverage information and mean concentration values for spring (MAM), summer (JJA), autumn (SON), and winter (DJF) are also presented in [Table S2](#).

The box-and-whisker plots in [Fig. 3](#) describe the variability of eBC mass concentrations in different regional sectors based on hourly averaged data from the relevant stations during the study period (2017–2019). Overall, the changes in concentrations reflect an increasing trend from north to south, with values ranging from 0.71  $\mu\text{g m}^{-3}$  (North) to 1.70  $\mu\text{g m}^{-3}$  (South). The relative contribution of different sources to these concentrations is discussed in [Section 3.2](#). The results revealed that eBC mass concentrations decreased in different sampling locations as follows: TR > UB > SUB > RB. Notably, IPR\_RB deviated from the observed trend, exhibiting high eBC concentrations ( $1.4 \pm 1.5 \mu\text{g m}^{-3}$ ), due to its location within the Po Valley PM hotspot ([Gilardoni et al., 2020](#); [Masiol et al., 2020](#); [Mousavi et al., 2019](#); [Sandrini et al., 2014](#)). The high concentrations at IPR\_RB emphasize the



**Fig. 3.** The variability of hourly averaged eBC mass concentrations at 50 sites between 2017 and 2019 categorized by the type of site and region. In the box and whisker plots, each box represents the range between the first and third quartile with the median value for each season represented by the inner line. The whiskers extend to the minimum and maximum values as determined by the 10th and 90th percentiles.

significant influence of meteorological conditions on eBC mass concentrations, as these readings are considerably higher than those at other UB or TR sites in Central Europe.

Furthermore, a decreasing trend in mass concentrations in the UB sites was observed moving from the Western and South-Western regions to Central and Northern Europe with the highest eBC mass concentrations reported for MLN\_UB ( $2.5 \pm 2.5 \mu\text{g m}^{-3}$ ) and GRA\_UB ( $1.9 \pm 2.0 \mu\text{g m}^{-3}$ ). At TR sites, lower concentrations are observed in the Northern regions compared to Central Europe. The results showed that there were significant disparities in eBC concentrations between the sites, with the highest eBC mass concentrations reported for MLN\_TR2 ( $3.4 \pm 2.3 \mu\text{g m}^{-3}$ ) and LND\_TR ( $2.9 \pm 2.2 \mu\text{g m}^{-3}$ ).

The absence of eBC measurements from Eastern Europe and the lack of measurements from TR sites in Southern Europe are clearly shown in Fig. 3. These limitations may affect the comprehensive assessment of spatiotemporal variability of eBC mass concentrations across Europe. Our primary focus in this study is to identify similarities and differences between the different European sites, rather than one-site peculiarities, which will be examined and discussed in detail in this section. As documented in prior studies on European aerosol phenomenology, meteorology, and regional geography can lead to significant differences in concentrations of pollutants such as eBC (Cavalli et al., 2016; Putaud et al., 2004; Van Dingenen et al., 2004). The greatest site-by-site variability in eBC mass concentrations for both the UB and TR sites was observed in Central Europe. The mean eBC concentrations at the TR sites ( $1.6 \pm 1.2 \mu\text{g m}^{-3}$ ) between 2017 and 2019 were higher than those measured at UB sites ( $1.3 \pm 1.4 \mu\text{g m}^{-3}$ ). The lowest mean concentration observed among the TR sites was  $0.67 \pm 0.68 \mu\text{g m}^{-3}$  in HEL\_TR6, while the highest was  $3.4 \pm 2.3 \mu\text{g m}^{-3}$  in MLN\_TR2. The eBC mass

concentration in MLN\_TR2 and LND\_TR were relatively similar, while MLN\_UB exhibited elevated eBC levels. In contrast, the eBC mass concentration at LND\_UB was significantly lower. The LND\_TR monitoring station is located on the kerbside of a street canyon (Marylebone Road), which is an area of the city that experiences heavy traffic (Rivas et al., 2020). Thus, local traffic was the main source of air pollution on this site (Ciupek et al., 2021). Similarly, because of its specific geographic location within the Po Valley (with extremely low winds and frequent atmospheric stagnation episodes), Milan is recognized as a regional pollution hotspot in Europe (Mousavi et al., 2019; Sandrini et al., 2014). Consequently, it has been characterized as a high-density polluted urban area dominated by traffic and biomass-burning sources that have been previously associated with elevated eBC mass concentrations (Invernizzi et al., 2011; Mousavi et al., 2019). In PAR\_TR (located in Paris, the second largest city in Europe), the mass concentration of eBC ( $2.5 \pm 1.7 \mu\text{g m}^{-3}$ ) is significantly impacted by primary emission sources, particularly traffic (Crippa et al., 2013; Favez et al., 2009). Conversely, the contribution of biomass-burning to eBC is relatively low at this station (Laborde et al., 2013).

In general, the recorded variations at UB sites ranged from a low of  $0.3 \pm 2.9 \mu\text{g m}^{-3}$  at STH\_UB to a high of  $2.5 \pm 2.5 \mu\text{g m}^{-3}$  at MLN\_UB. GRA\_UB also exhibited high eBC mass concentrations ( $1.9 \pm 2.0 \mu\text{g m}^{-3}$ ), likely due to its location in a narrow valley, as well as the open burning of wood and strong and frequent thermal inversion during the cold months (Titos et al., 2017). Among the SUB sites, the highest eBC mass concentrations were observed at BUC\_SUB ( $2.1 \pm 3.0 \mu\text{g m}^{-3}$ ), primarily due to the influence of noticeable biomass-burning during summer (Chen et al., 2022). The lowest eBC mass concentration among all sites was reported at SMR\_RB ( $0.2 \pm 0.20 \mu\text{g m}^{-3}$ ) and STH\_UB ( $0.3$



$\pm 0.3 \mu\text{g m}^{-3}$ ). STH\_UB is affected by wood smoke emissions during wintertime (Krecl et al., 2011), while SMR\_RB—a rural station—typically experiences one of the lowest eBC loads in Europe (Hyvärinen et al., 2011; Yttri et al., 2011). The ranges of eBC mass concentrations described above also reflect the diverse environmental conditions across Europe, including variations in meteorology and urban and regional emissions patterns (Bressi et al., 2021).

In the context of eBC mass concentrations within specific regions, a relatively low degree of site-by-site variability was observed at TR sites in the Northern countries. Comparatively, the regions of Western (Netherlands) and Northern (Finland) Europe exhibited noticeably lower eBC mass concentrations compared to other urban areas across Europe. Finland's geographical isolation from the more densely populated regions of Europe due to their separation by the Baltic Sea results in the reduced influence of long-range pollution originating from the more polluted areas (Luoma et al., 2021). In Northern Europe, biomass-burning emissions have been identified as a significant source of air pollutants (Helin et al., 2018; Hienola et al., 2013; Hyvärinen et al., 2011; Luoma et al., 2021). Our results showed that the higher eBC concentrations observed at the Northern SUB sites are consistent with previous studies that have highlighted increased eBC contributions from residential wood and other biomass combustion during the cold seasons in the SUB area (Helin et al., 2018; Luoma et al., 2021). In general, low site-by-site variability was observed in both SUB and RB sites in Northern countries. Northern countries can thus be characterized by their low eBC concentrations and site-by-site variability.

The variability of eBC concentrations at Western European TR sites was found to be higher than at UB sites. Schaap and Denier van der Gon (2007) previously demonstrated that TR stations in the Netherlands exhibited greater variability in eBC concentrations compared to UB and RB sites. For instance, the relatively high eBC mass concentrations in Rotterdam (NLD\_TR3;  $1.7 \pm 1.2 \mu\text{g m}^{-3}$ ) were mainly attributed to high road traffic emissions (Keuken et al., 2009).

Data was only available at three stations in South-Eastern and Eastern European countries, limiting the efficacy of site-by-site variability analyses in these particular regions. However, these stations can still be compared to other EU regions. Notably, BUC\_SUB ( $2.1 \pm 3.0 \mu\text{g m}^{-3}$ ) reported significantly higher eBC mass concentrations compared to similar sites in other regions. ATH\_SUB is affected by both traffic and residential and commercial combustion sources (Diapouli et al., 2017). However, the eBC concentrations at ATH\_UB ( $1.7 \pm 2.2 \mu\text{g m}^{-3}$ ) were comparable to other UB sites in Southern Europe and exhibit values that are nearly double than those observed at ATH\_SUB ( $0.80 \pm 0.5 \mu\text{g m}^{-3}$ ), suggesting that there is significant spatial variability in ambient eBC and its emission sources in Athens (Grivas et al., 2019; Kaskaoutis et al., 2021).

In South-Western Europe, GRA\_UB exhibited higher eBC mass concentrations compared to other sites, potentially influenced by meteorological patterns, orographic factors, and high biomass-burning emissions (Titos et al., 2017; Van Drooge et al., 2022). The eBC mass concentrations at VLC\_UB, BCN\_UB, and MAR\_UB were similar, while the lowest eBC mass concentration was recorded at MAD\_UB due to the least influence from traffic in this site. However, studying the variability of long-term measurements of eBC mass concentrations at the European scale is challenging since the characterization of eBC mass concentration by station is restricted due to significant regional variability.

It should be noted that intense desert dust episodes (DDE) can also contribute to the measured absorption and can thus affect the calculated eBC concentrations. The contribution of this dust to absorption measurements has been most commonly observed at remote sites where eBC concentrations are usually low (e.g., Ripoll et al., 2015). However, at urban sites where eBC is expected to dominate absorption, the impact of DDE on eBC concentrations is relatively insignificant, as can be seen by the relationship between eBC and  $\text{NO}_2$  concentrations at BCN\_UB over three years of data (see Fig. S2). It should also be noted that DDE events

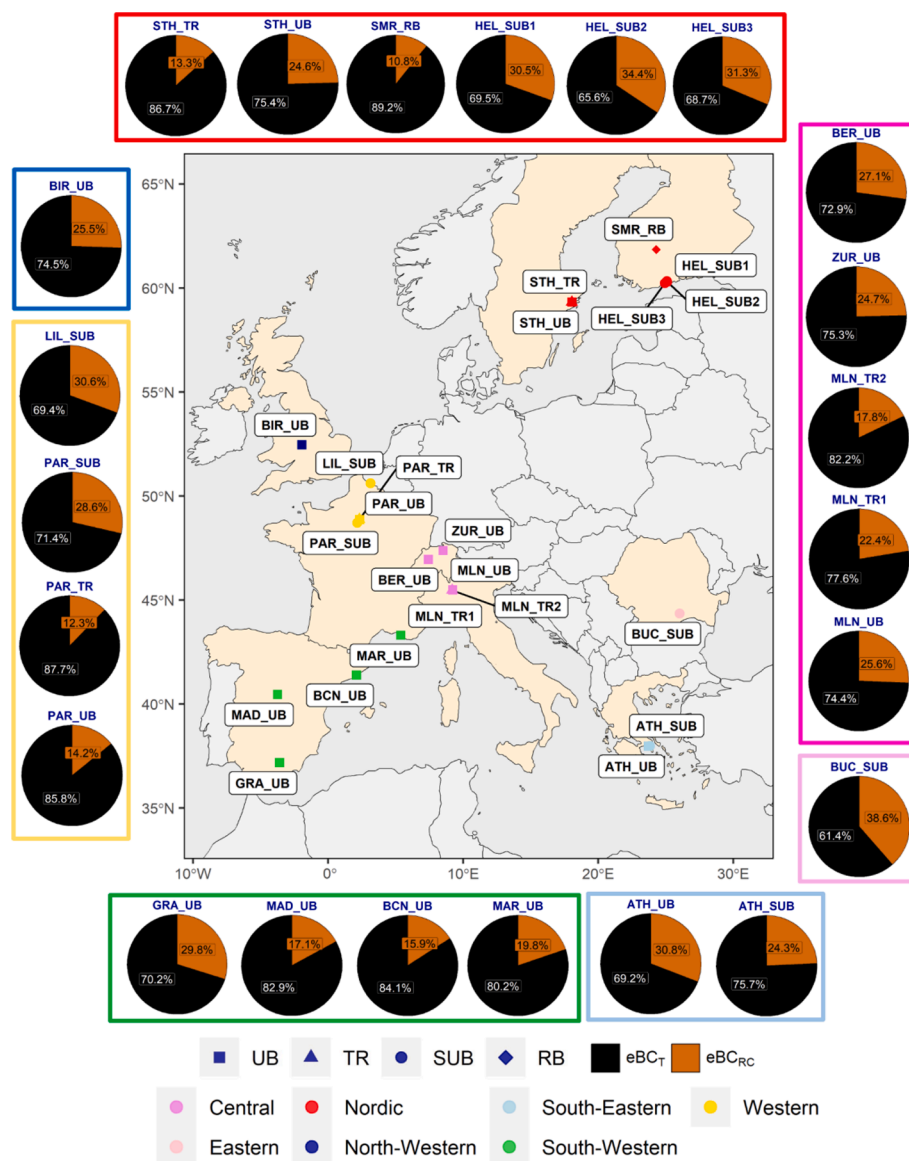
are sporadic, thus further reducing the influence of DDE on urban sites over long timescales such as those used in this study.

### 3.2. Source apportionment analysis of eBC

The aethalometer model was applied to 23 AE33 datasets at seven wavelengths; the results of this analysis are presented here. This model was used to determine the relative contribution of  $\text{eBC}_{\text{RC}}$  and  $\text{eBC}_{\text{T}}$  to bulk eBC based on the particle light absorption coefficients calculated over the entire study period (2017–2019). The mean concentrations of  $\text{eBC}_{\text{T}}$  and  $\text{eBC}_{\text{RC}}$ , as well as their corresponding standard deviations, were determined using hourly averaged data at different sites during the study period and are reported in Table S3 (using  $\text{AAE}_{\text{T}} = 1$  and  $\text{AAE}_{\text{RC}} = 2$ ) and Table S4 (using  $\text{AAE}_{\text{T}} = 0.90$  and  $\text{AAE}_{\text{RC}} = 1.68$ ). Fig. 4 shows the results of the aethalometer source apportionment model in Europe for  $\text{AAE}_{\text{T}} = 1$  and  $\text{AAE}_{\text{RC}} = 2$  (Sandradewi et al., 2008a). It shows the regional variations in the relative contributions of  $\text{eBC}_{\text{T}}$  and  $\text{eBC}_{\text{RC}}$  with an increasing trend in the relative contribution of  $\text{eBC}_{\text{T}}$  from Northern to Central, Western, and South-Western Europe. The outcomes of the source apportionment analysis are summarized in Fig. 5, which provides a comparison of the relative contributions of  $\text{eBC}_{\text{T}}$  and  $\text{eBC}_{\text{RC}}$  between the TR and the UB/SUB sites using stacked histograms; the error bars indicate the respective variances of the two sources. These comparisons are presented using pie charts in Fig. 4. It shows that  $\text{eBC}_{\text{T}}$  had consistently higher relative contributions compared to  $\text{eBC}_{\text{RC}}$ . The highest relative contribution of  $\text{eBC}_{\text{T}}$  was obtained in PAR\_TR (88%), STH-TR (87%), and MLN\_TR (82%) sites. However, there were some regions and sites at which  $\text{eBC}_{\text{RC}}$  contributions were also relatively high.

Previous research has shown that eBC mass concentrations are predominantly influenced by residential and commercial combustion sources—particularly wood burning—during heating seasons (Crilley et al., 2015; Herich et al., 2011; Zhang et al., 2020a, 2020b, 2020c). The greatest relative  $\text{eBC}_{\text{RC}}$  contribution was observed at BUC\_SUB (39%), the HEL\_SUB sites (30–34%), GRA\_UB (30%), BER\_UB (27%), and MLN\_UB (26%). The lowest  $\text{eBC}_{\text{T}}$  contributions were observed in Eastern Europe, while the highest contributions were recorded in Central and Western Europe (88%, 82%, and 74% at PAR\_TR, MLN\_TR2, and MLN\_UB, respectively). It is worth noting that the relative contribution of  $\text{eBC}_{\text{T}}$  in the Northern TR sites was much lower compared to TR sites in Western Europe, such as PAR\_TR, with the exception of the UB and TR sites of MLN in Central Europe. In contrast, in South-Eastern Europe, both ATH\_UB and ATH\_SUB exhibited higher relative contributions of  $\text{eBC}_{\text{T}}$  despite being recognized as hot spots for residential wood burning, especially during winter (Kaskaoutis et al., 2021; Liakakou et al., 2020). There was a clear decreasing trend in  $\text{eBC}_{\text{RC}}$  contributions from Northern to Central and Southern Europe. In Central, Eastern, and Northern Europe, the relative contribution of  $\text{eBC}_{\text{RC}}$  to eBC was found to be higher compared to other regions (39% in BUC\_SUB, 30–34% in HEL\_SUB sites, and 26% in MLN\_UB). This pattern may be due to the increased emissions of solid fuel from residential areas during the cold season in these particular regions.

The higher relative  $\text{eBC}_{\text{RC}}$  contributions in South-Eastern Europe (ATH sites) during winter are likely attributed to the burning of wood for heating purposes (ATH\_UB 31%, ATH\_SUB 24%). Consequently, the ratio of biomass to fossil-fuel combustion emissions in this region was higher in winter, particularly on weekends (Kaskaoutis et al., 2021; Merabet et al., 2019). The SMR\_RB station is mainly affected by regionally sourced and long-range transported pollutants in Northern Europe (Hyvärinen et al., 2011). In addition, the SMR\_RB station may be affected by the surrounding station buildings as well as the city of Tampere itself (Yttri et al., 2011). In the Helsinki region, stations HEL\_SUB1 to HEL\_SUB3 are located in detached housing areas that are affected by the utilization of fireplaces for wood burning in winter (32% relative contribution to  $\text{eBC}_{\text{RC}}$ ) in addition to road traffic emissions (Helin et al., 2018; Fung et al., 2022). STH\_TR, located in the city center, is heavily influenced by traffic emissions (75%  $\text{eBC}_{\text{T}}$ ) (Krecl et al., 2011;



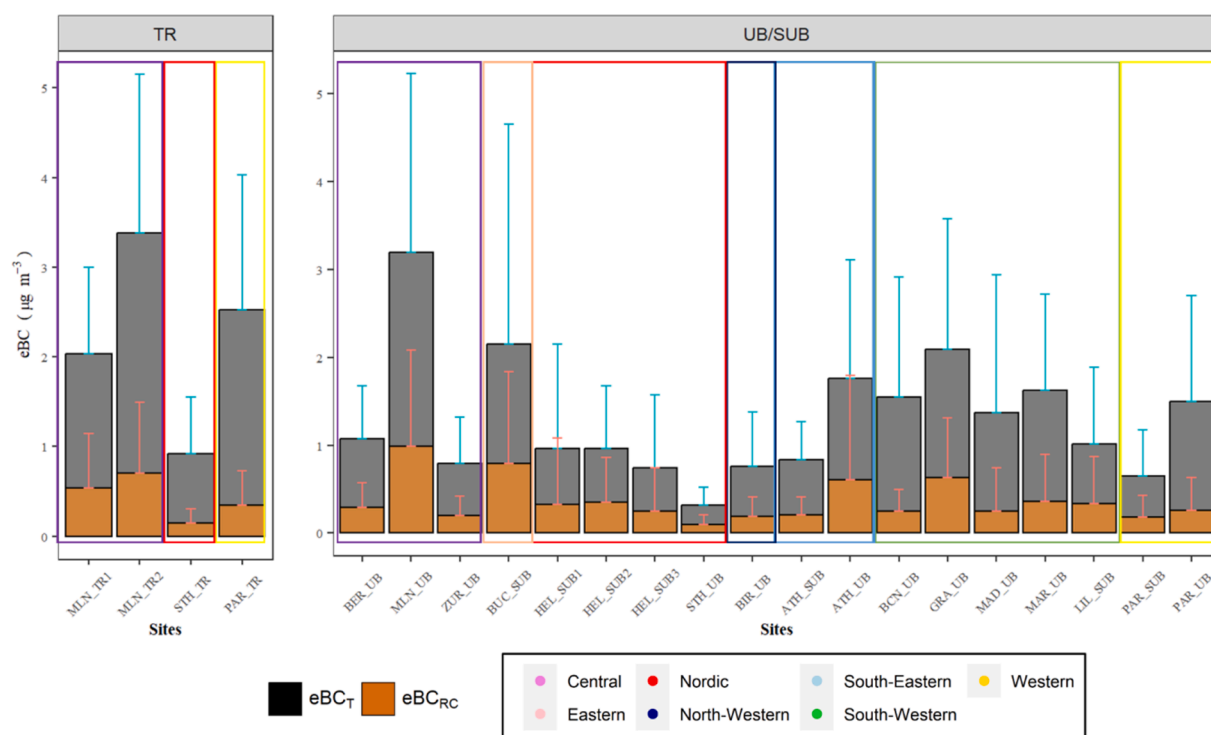
**Fig. 4.** The results of the source apportionment analysis of eBC mass concentrations for 23 European sites used over the study period (2017–2019). The aethalometer model was applied using ( $AAE_T = 1$ ) for traffic emission sources and ( $AAE_{RC} = 2$ ) for residential and commercial emission sources. The colors highlight the geographic locations of the sites that supplied AE33 eBC data.

2017).

Of the South-Western UB sites, high contributions of eBC<sub>T</sub> and eBC<sub>RC</sub> were observed in BCN\_UB (84%) and GRA\_UB (30%), respectively. Similarly, MAD\_UB (83%) and MAR\_UB (80%) also exhibited high relative contributions of eBC<sub>T</sub>, consistent with findings reported by Baldasano (2020). MAR\_UB is located in the most important Mediterranean seaport and is consequently influenced by a substantial amount of industrial urban emissions (Chazeau et al., 2022; El Haddad et al., 2013; Salameh et al., 2018). In Western sites, high eBC<sub>T</sub> and eBC<sub>RC</sub> contributions were observed in sites that are influenced by heavy traffic and biomass burning, such as PAR\_TR (88%) and LIL\_SUB (31%), respectively. In particular, LIL\_SUB is highly influenced by biomass-burning emissions during spring, autumn, and winter as previously reported by Cordell et al. (2016). A recent study conducted at the LIL\_SUB site revealed that, during the winter season, a significant portion of the organic aerosols consisted of less oxidized fractions (LO-OOA), and has been identified as originating from aged biomass burning. The study suggests that approximately half of the organic aerosols generated during winter can be attributed to wood combustion (Chebaicheb et al.,

2023). In North-Western Europe, BIR\_UB exhibited a dominant contribution of eBC<sub>T</sub> (74%) due to its higher traffic emissions compared to domestic heating; this was consistent with a previous study on the variations and level of the eBC<sub>T</sub> contribution at BIR\_UB by Singh et al. (2018). In general, the relative contribution of eBC<sub>T</sub> and eBC<sub>RC</sub> is influenced by the geographical location and the prevailing type of combustion source at each station.

Moreover, we performed a comparison of the relative contributions of eBC<sub>RC</sub> and eBC<sub>T</sub> using two sets of AAE values: 1 and 2, and the AAE values proposed by Zotter et al. (2017), as shown in Table S4. In general, the relative contribution of residential and commercial sources to eBC increased by 25–45% when using the Zotter values (see Fig. S3). It should be noted that the AAE values proposed by Zotter et al. (2017), were determined at Swiss sites that were strongly influenced by biomass-burning emissions. However, based on the available data, we cannot definitively conclude which AAE pair (1, 2 vs. 0.90, 1.68) was more appropriate for the sites considered in our study. It is reasonable to apply the Zotter AAE values at sites with similar characteristics to those used in Zotter et al. (2017). However, there are some concerns regarding the



**Fig. 5.** The source apportionment analysis results of TR (traffic) vs UB/SUB (urban/suburban) sites. Stacked histograms with error bars were used to present the variance associated with the two sources based on geographic regions.

extensive use of Zotter TR values. For instance, in Barcelona, the Zotter values result in an excessively high contribution of  $eBC_{RC}$  to  $eBC$  (41% in winter), which contradicts recent studies demonstrating the very low contribution of biomass-burning to OA in this city (4–6% in winter; [Via et al. 2021](#)).

It thus cannot be assumed that the AAE values remain constant across all sites. To minimize these uncertainties, the use of site-specific AAE values is recommended; alternatively, a range of robust and consistent AAE values that accounts for the variability observed in the specific study area should be used. There are studies that have recommended the use of AAE frequency distributions as an alternative method to estimate site-specific AAE values ([Tobler et al., 2021](#)). Alternatively, the incorporation of time-dependent AAEs into the aethalometer model can be proposed, with the aim of evaluating its performance. In the next section, trends in  $eBC_T$  and  $eBC_{RC}$  will be analyzed to determine if there are stable patterns for  $eBC_{RC}$  and  $eBC_T$  in the data by considering monthly-deseasonalized average values for larger data series.

### 3.3. Seasonal variability

#### 3.3.1. $eBC$ seasonal pattern

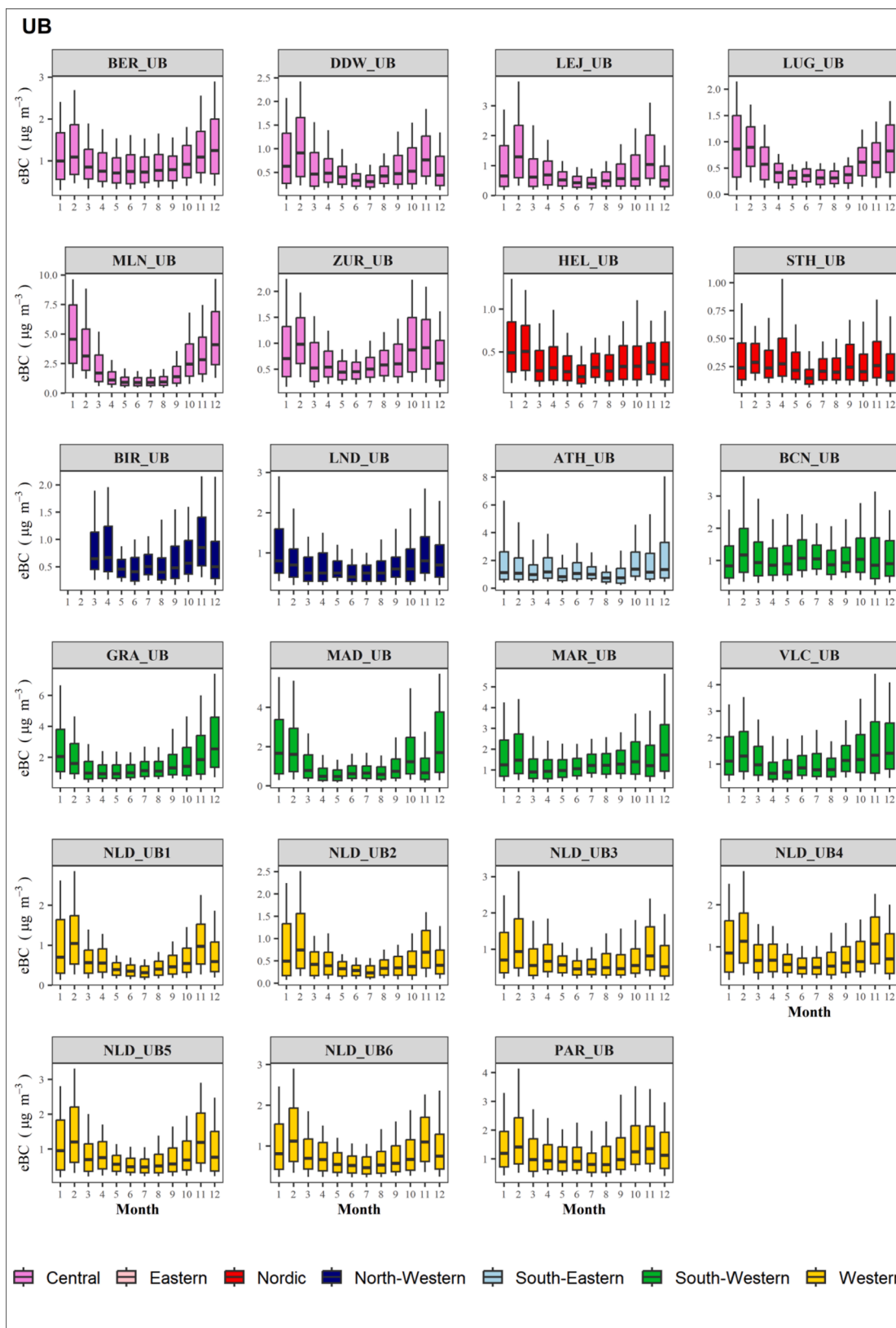
[Fig. 6](#) shows the average monthly variation in the total  $eBC$  mass concentrations across all sites between 2017 and 2019. In cases where monitoring sites provided  $eBC$  observations from multiple instruments, such as MAAP, AE31, and AE33, the analysis in this section primarily considers the seasonal variability observed from MAAP data. The priority in the use of specific instrument data was as follows: MAAP > AE33 > AE31 > AE22. The  $eBC$  mass concentrations were noticeably higher during winter (DJF) at a majority of sites, which may be attributed to the seasonal variability of  $eBC$  emission sources and meteorological conditions. In general, winter periods are characterized by lower mixing layer heights, lower wind speeds, and enhanced atmospheric stability, which contribute to the greater accumulation of pollutants, even in the presence of frequent precipitation ([Cassée et al., 2019](#)). In addition, higher winter domestic heating emissions contribute to

increased  $eBC$  ([Minderyté et al., 2022](#)). The trend of higher  $eBC$  concentrations during winter has been already described in numerous studies ([Becerril-Valle et al., 2017](#); [Herich et al., 2011](#); [Jing et al., 2019](#); [Lyamani et al., 2011](#); [Merabet et al., 2019](#); [Pandolfi et al., 2014](#); [Titos et al., 2017](#)). The observed high winter  $eBC$  patterns are consistent with similar patterns of high winter  $NO_x$  and CO reported in many European and US studies, as both of these tracers are also emitted in large quantities by road traffic ([Grange et al., 2021](#); [Helin et al., 2018](#); [Karagulian et al., 2015](#); [Mousavi et al., 2018](#)).

There are some cities where this seasonal cycle is not as pronounced or even non-existent. For instance, at BCN\_UB, the effect of atmospheric inversion is relatively weak and biomass-burning in winter only has a minor contribution to ambient PM levels ([Amato et al., 2016](#)). PM speciation studies conducted at BCN\_UB revealed that levels of biomass-burning organic aerosol in winter were lower there than in other European sites ([Via et al., 2021](#)).

Seasonal variations in  $eBC$  mass concentration were relatively stable in several Northern European sites, particularly at HEL\_TR1, HEL\_TR2, and STH\_TR. These sites all are located in Nordic street canyons, where local traffic is the dominant source of  $eBC$  throughout the year, and there is no significant contribution from local wood combustion at these sites ([Krecl et al., 2011](#); [Helin et al., 2018](#)). In addition, the low mean height of the boundary layer and low wind speeds during the early summer morning rush hours leads to elevated  $eBC$  mass concentrations in Helsinki during summer ([Barreira et al., 2021](#)). Seasonal variations at the STH\_TR site can also be affected by the abatement of traffic since traffic exhaust has been found the most important source in city of Stockholm ([Segersson et al., 2017](#)).

The UB and TR sites in Europe, predominantly concentrated in the Western and Northern regions, exhibit a less pronounced seasonal variability, particularly in the Northern countries. In contrast, both UB and TR sites exhibited pronounced high winter patterns in Central Europe. The spatial variability observed at the SUB and RB sites was further influenced by their geographic locations and their local climate/meteorology.



**Fig. 6.** The averaged monthly variability of eBC mass concentrations based on the type of site and region between 2017 and 2019. In box plots, the range of the box depicts the bounds of the 25th and 75th percentile of the data. The whiskers extend to the minimum and maximum values, determined by the 10th and 90th percentiles. The bold horizontal inner line shows the median for each month. The shaded background colors in box plots represent geographic locations.

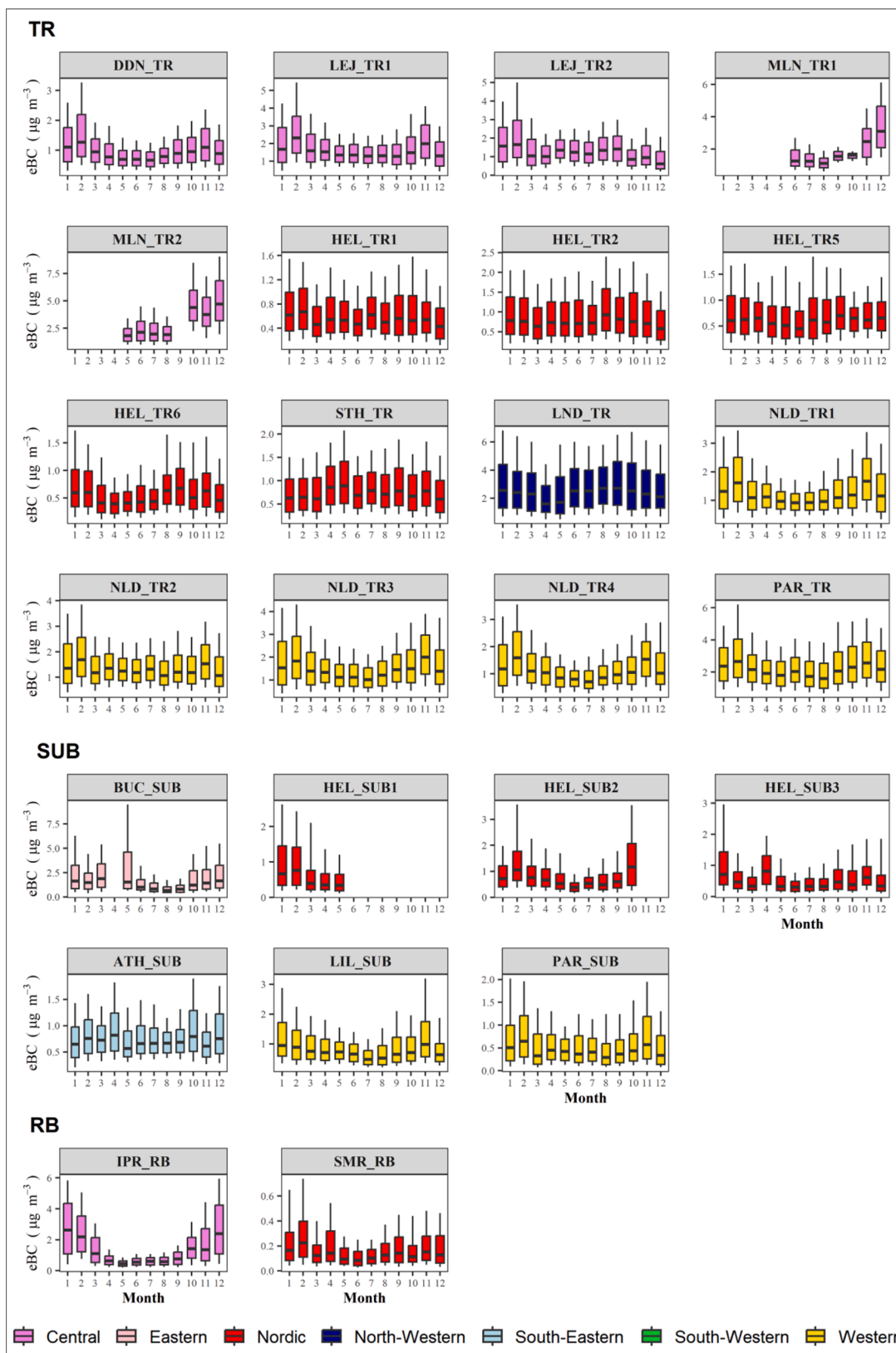
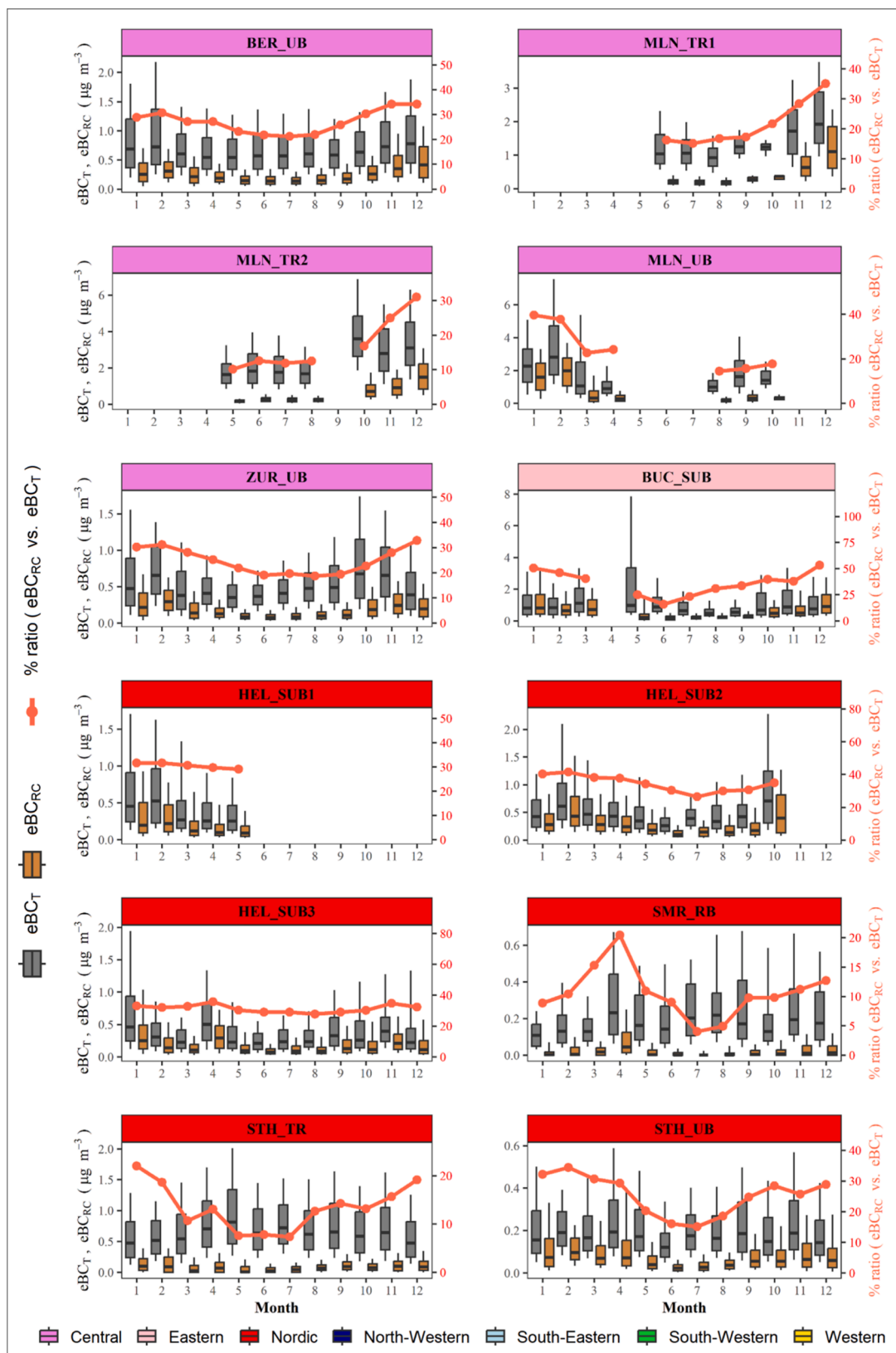


Fig. 6. (continued).

### 3.3.2. Seasonal trends in eBC<sub>T</sub> and eBC<sub>RC</sub>

The monthly variations of eBC<sub>T</sub> and eBC<sub>RC</sub> are presented in Fig. 7. In general, the results revealed that eBC source contributions exhibited pronounced seasonality and a significant inter-annual fluctuation during

the study period (2017–2019). This is supported by Fig. 7, which illustrates the high variability of eBC<sub>RC</sub>/eBC<sub>T</sub> ratios for all studied sites, which vary from 23 to 87% across Europe. These fluctuations may be a function of the distance of the site to its traffic, residential, and



**Fig. 7.** The averaged monthly variability of  $eBC_T$  and  $eBC_{RC}$  concentrations based on the type of sites and region between 2017 and 2019. In boxplots, each box represents the range between the first and third quartile with the median value for each season distribution represented by the inner line. The whiskers extend to the minimum and maximum values, determined by the 10th and 90th percentiles.

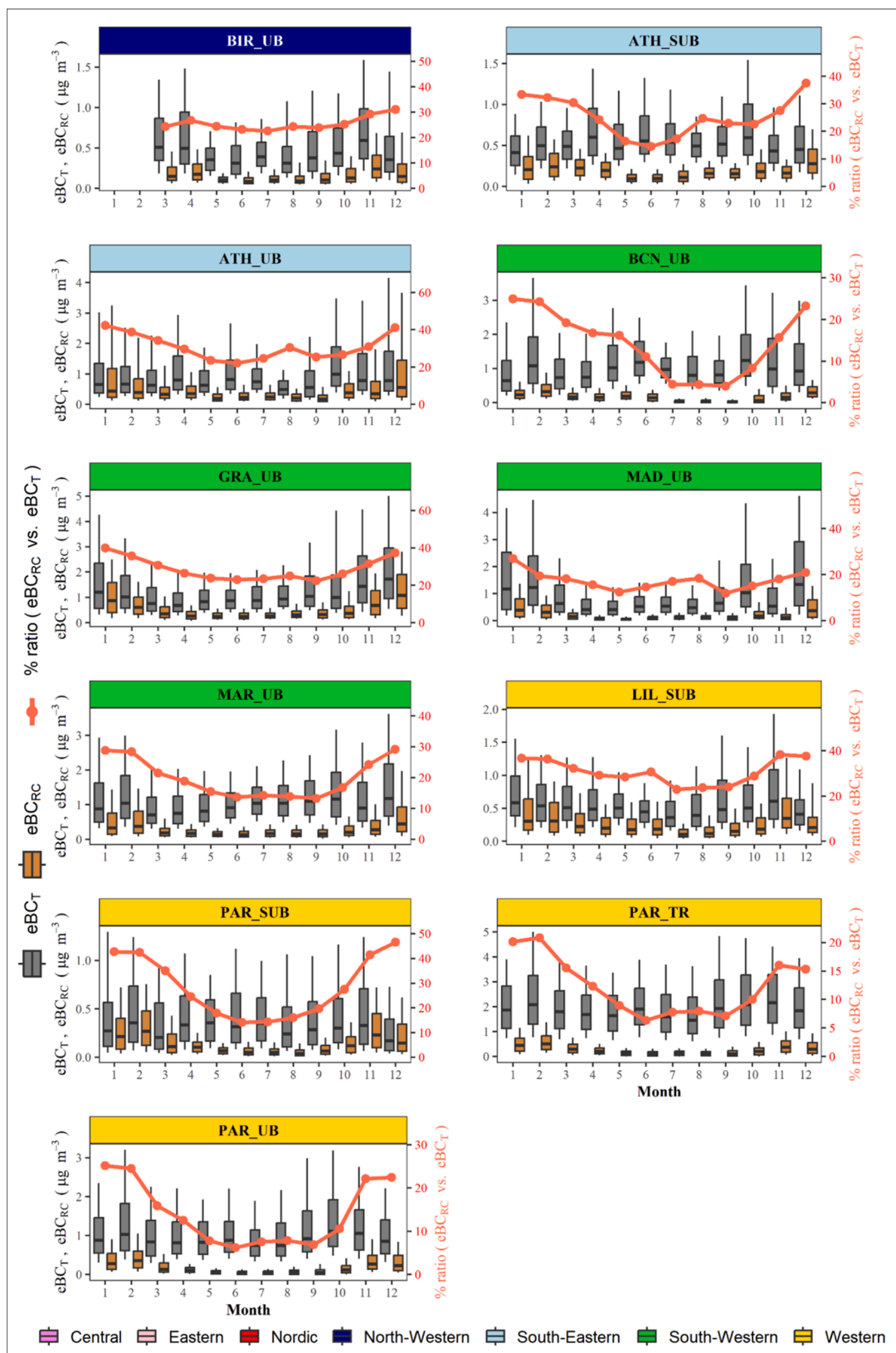


Fig. 7. (continued).

commercial source emissions, the relative influence of other emission sources, intra-annual variations in emissions from different combustion sources, and changes in meteorological conditions (Becerril-Valle et al., 2017; Bond et al., 2013; Helin et al., 2018; Kaskaoutis et al., 2021;

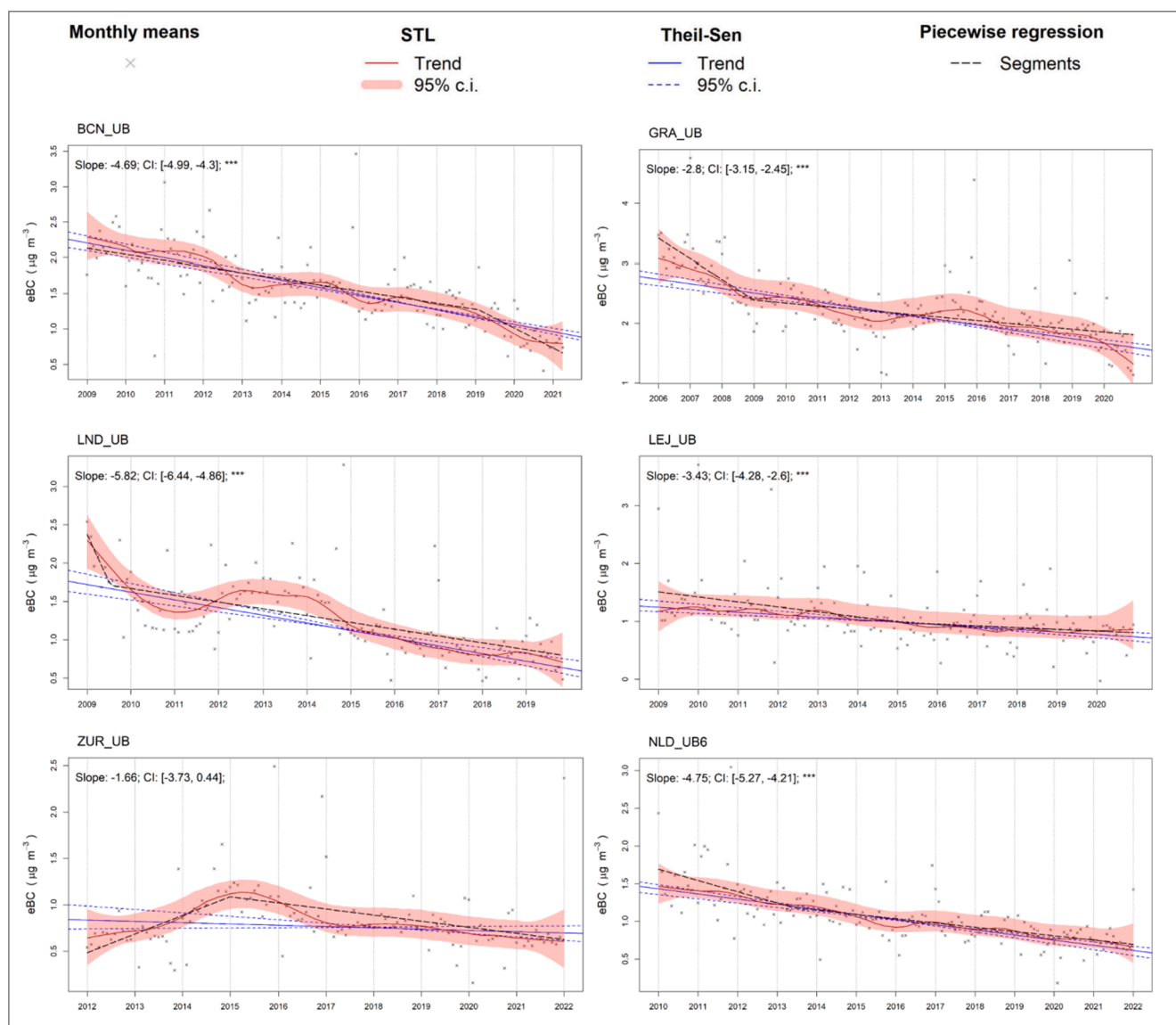
Luoma et al., 2021; Ziola et al., 2021).

These observations are consistent with the decrease in the ratio of  $eBC_T/eBC_{RC}$  emissions during winter and their corresponding increase during summer (Hopke et al., 2022; Rigler et al., 2020; Rivas et al.,

2021), a trend that was also observed in this study for most UB/TR sites. Conversely, the  $eBC_{RC}/eBC_T$  emission ratio was expected to exhibit the opposite pattern, decreasing during summer and increasing during winter (Fig. 7). The monthly variation and the relative contribution of  $eBC_T$  and  $eBC_{RC}$  sources across the different sites are also presented as a 100% stacked histogram (see Fig. S4), offering insights into the spatial distribution and dominance of specific source types. The values used for these histograms were determined through the application of the aethalometer source apportionment model at 23 different sites using  $AAE_T = 1$  and  $AAE_{RC} = 2$ . The y-axis of the histograms refers to the relative contribution of each source category (as a percentage), while the x-axis indicates their monthly variation. The stacked bars refer to the relative contributions of  $eBC_T$  and  $eBC_{RC}$  sources for each site.

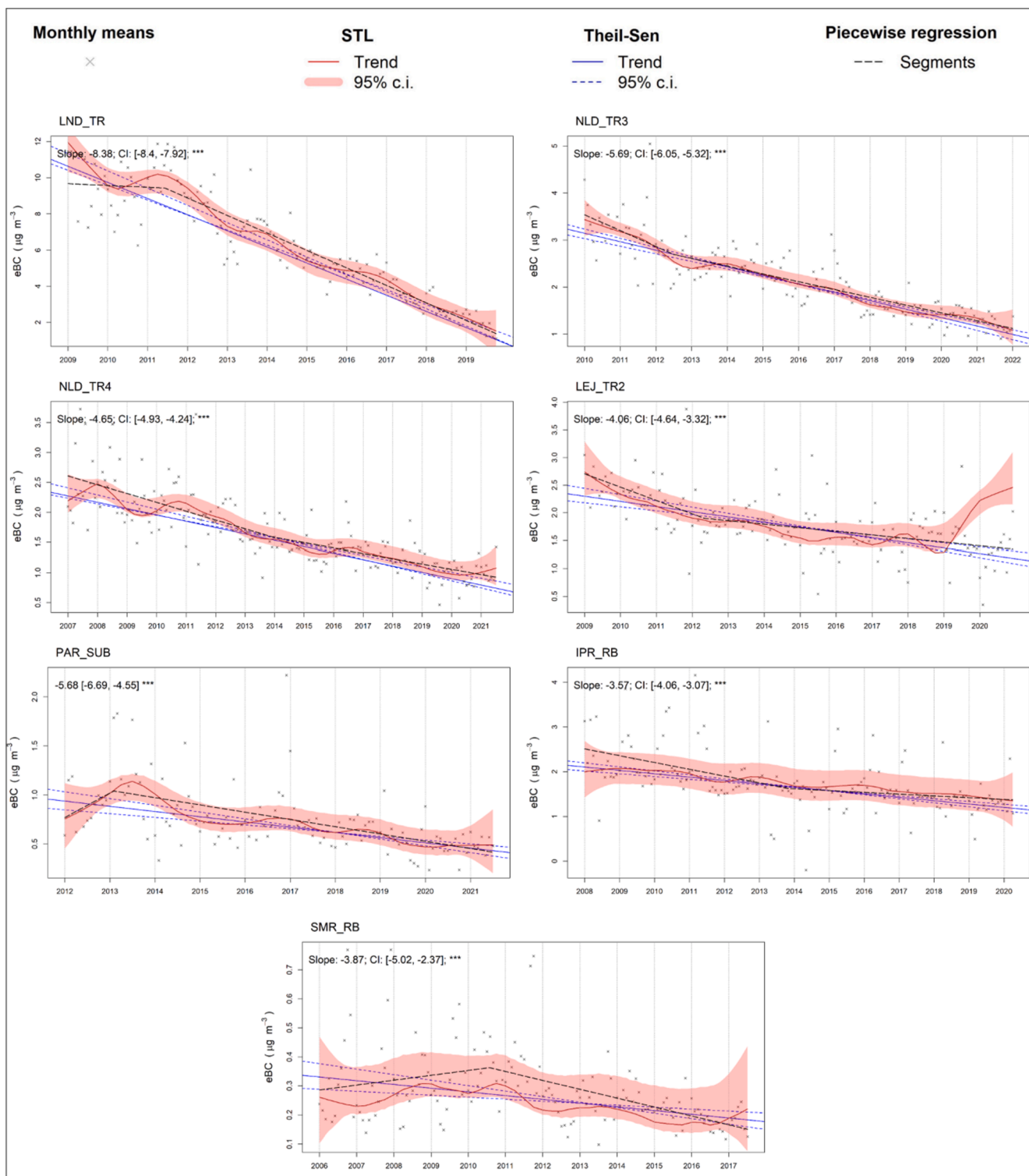
The relative  $eBC_T$  contributions increased during winter as a result of more stagnant atmospheric conditions and increased traffic emissions during this season. At the same time, there was an increase in  $eBC_{RC}$  contributions due to the emissions from domestic sources (e.g., heating). The seasonal variations in  $eBC_T$  were observed to be lower compared to  $eBC_{RC}$ ; this discrepancy is related to the lesser degree of seasonal dependence exhibited by urban traffic flow compared to the seasonality

of solid fuel combustions, such as biomass- and coal-burning, which are prevalent at most sites (Harrison et al., 2013; Martinsson et al., 2017). Furthermore, it is worth noting that residential heating activities can also contribute to local  $NO_x$  and eBC, such as at the detached housing areas around the HEL sites (Helin et al., 2018). In the context of seasonal variations, the contribution of  $eBC_T$  was at a maximum during winter ( $3.5 \mu\text{g m}^{-3}$ ) at MLN\_TR2, with notably elevated values also observed during the summer ( $2.0 \mu\text{g m}^{-3}$ ) at the same site. In contrast, the lowest values of  $eBC_T$  ( $0.22 \mu\text{g m}^{-3}$ ) were measured in both summer and winter at STH\_UB. The seasonal patterns of  $eBC_{RC}$  were similar, with the maximum concentrations observed at MLN\_UB ( $1.90 \mu\text{g m}^{-3}$ ) and MLN\_TR2 ( $1.65 \mu\text{g m}^{-3}$ ) during the winter and the minimum concentrations observed at STH\_UB ( $0.22 \mu\text{g m}^{-3}$ ). In summer, GRA\_UB ( $0.34 \mu\text{g m}^{-3}$ ) and ATH\_UB ( $0.30 \mu\text{g m}^{-3}$ ) exhibited the maximum contribution of  $eBC_{RC}$ . Seasonal patterns of  $eBC_T$  and  $eBC_{RC}$  were not homogeneous across the studied sites, possibly due to variations in the emission and climate patterns in the different regions and cities.



**Fig. 8.** Trends in eBC mass concentration for selected UB sites. The slope of the trends is given in  $\% \text{yr}^{-1}$ . The statistically significant trends of each site are represented by \*\*\* for  $p < 0.001$ , \*\* for  $p < 0.01$ , \* for  $p < 0.05$ , + for  $p < 0.1$ , and ns for not significant. The 95% confidence intervals (CI) for STL and MK-TS analyses were computed by bootstrapping.





**Fig. 9.** Trends in eBC mass concentration for selected TR and RB sites. The slope of the trends is given in % yr<sup>-1</sup>. The statistically significant trends of each site are represented by \*\*\* for  $p < 0.001$ , \*\* for  $p < 0.01$ , \* for  $p < 0.05$ , + for  $p < 0.1$ , and ns for not significant. The 95% confidence intervals (CI) for STL and MK-TS analyses were computed by bootstrap.

### 3.4. Trend analysis

Figs. 8 and 9 represent the estimated inter-annual trends of eBC mass concentrations for selected sites with more than nine years of eBC observations between 2006 and 2020. The results of the Theil-Sen trend analysis, which include the slopes and corresponding confidence intervals, are presented in Fig. S6. This figure presents the standard errors

of the breakpoints using the “segmented” R package, as well as the slopes and confidence intervals obtained from the Theil-Sen trend analysis. Annual deseasonalized STL trends and MK-TS, Theil-Sen slopes as well as the results of the piecewise linear regression computed throughout the year (annual) can be found in Table S5 and S6. Table S5 provides the corresponding  $p$ -values, slopes, and confidence intervals (CI). Table S6 provides details on the number of breakpoints, the dates of

identified breakpoints, standard errors, adjusted  $R^2$  values (model accuracy), root mean squared errors (RMSEs), and mean absolute errors (MAEs) expressed in  $\mu\text{g m}^{-3}$ .

A consistent, statistically significant decreasing trend was detected at all sites during the observation period. This decreasing eBC trend is consistent with several European and American studies that have been conducted in recent years (Ahmed et al., 2014; Collaud Coen et al., 2020; Jafar and Harrison, 2021; Kutzner et al., 2018; Luoma et al., 2021; Masiol et al., 2018; Mousavi et al., 2018; Olstrup et al., 2018; Sun et al., 2021; Veld et al., 2021; Wang et al., 2014). These results are in contrast to previous studies conducted between 1960 and 2007, which reported an increase in eBC, especially in developing countries (Wang et al., 2014). In this study, the observed decreasing trends were associated with the European- and local-scale measures to improve AQ in urban areas. The observed trends in eBC mass concentrations can be influenced by different factors specific to each site over time, the details of which are beyond the scope of this paper. However, it is important to note that the enforcement of diesel filter traps since the EURO5 vehicle emission standards were enacted and the economic crisis in 2008 may have had a decisive contribution to these trends (Lyamani et al., 2011; Querol et al., 2014). A summary of the results obtained by applying three methods for the trend analyses is presented in Figs. 8 and 9.

In North-Western Europe, statistically significant decreasing trends were identified at both LND\_TR and LND\_UB sites between 2009 and 2019, with annual percentage decreases of  $-5.8\%$  and  $-8.4\%$   $\text{yr}^{-1}$ , respectively. Our findings are consistent with the work published by Ciupek et al. (2021), who also reported a decreasing trend in eBC concentrations for LND\_TR and LND\_UB sites between 2014 and 2019, with decreases of up to  $-13\%$ . These reductions have been attributed to the decline in PM emissions from diesel vehicles over the last decade, as well as an increase in the proportion of low-emission buses in London (Ciupek et al., 2021). The results of piecewise linear regression analysis confirmed that this strong drop had a detectable breakpoint in 2009 at LND\_UB. A similar breakpoint was also observed at LND\_TR in 2011. These findings are consistent with the trend analyses of eBC concentrations conducted in the UK between 2009 and 2016, particularly highlighting the substantial decrease in eBC of around  $-8 \pm 3\%$  per year observed at LND\_TR (Singh et al., 2018). It should be noticed that the declining trend in LND\_TR has been reported to be notable compared to other sites in UK (Singh et al., 2018). The findings of Singh et al. (2018) and Jafar and Harrison (2021) emphasize the fact that the reduced eBC levels at both London sites were due to the decrease in vehicle exhaust emissions and the effect of traffic emission abatement policies.

In Central Europe, the decreasing trends in eBC mass concentrations over time were more pronounced at German TR sites compared to UB sites. This trend is consistent with recent literature that found that trends at TR sites show larger declines in eBC concentrations over time compared to UB sites (Kutzner et al., 2018). Decreasing trends were observed at LEJ\_UB and LEJ\_TR sites in Central Europe, with annual percentage decreases of  $-3.4\%$   $\text{yr}^{-1}$  and  $-4.1\%$   $\text{yr}^{-1}$  respectively (see Figs. 8 and 9). These results are consistent with a recent study that demonstrated a decreasing trend in eBC over Central Europe between 2009 and 2018 (Sun et al., 2020, 2021). The introduction of low emission zones in Leipzig in 2011 has also led to substantial reductions in eBC mass concentrations, resulting in an 18–36% decrease between 2005 and 2014 (Kutzner et al., 2018; Rasch et al., 2013). Piecewise linear regression also revealed a strong drop with one breakpoint at LEJ\_TR2 in 2012. However, STL and piecewise regressions did not identify any similar rapidly declining trends in eBC mass concentrations around the UB site. In addition, a decreasing trend of  $-3.5\%$   $\text{yr}^{-1}$  was observed at IPR\_RB. Piecewise linear regression also revealed one breakpoint in 2014. According to the published report in 2018, emissions stemming from biomass burning for domestic heating and fossil fuel combustion related to transport and domestic heating around IPR\_RB showed an increase of eBC<sub>RC</sub>/eBC<sub>T</sub> ratio during the years 2009–2010 that could potentially be linked to the economic crisis. Then

remained relatively stable until 2013 (with a local minimum in 2011), and have generally exhibited a downward trend since that time (Putaud et al., 2018).

This study also revealed a statistically significant decreasing trend in eBC of  $-4.7\%$   $\text{yr}^{-1}$  between 2010 and 2020 at BCN\_UB, consistent with a reported significant decrease ( $-18\%$ ) in eBC from 2014 to 2018 (Via et al., 2021). Piecewise linear regression and STL showed best fits with the Theil-Sen slope with one breakpoint in 2019. This suggests that the main driver of this decrease in mass concentrations may have been the implementation of traffic emission abatement policies (Carnerero et al., 2021). The GRA\_UB site exhibits a slightly weaker decreasing trend, although it is still statistically significant ( $-2.8\%$   $\text{yr}^{-1}$ ; see Fig. 8). The results of the STL and piecewise linear regression analysis are consistent with Theil-Sen, with one significant breakpoint detected in 2009, probably related with the economic recession experienced in 2008. This observation may also have been strongly affected by the implementation of EURO 4 and EURO 5 standards, which led to a reduction in emissions of PM and CO from both road traffic (resulting in a progressive downward trend) and industrial sources (resulting in sharp changes in 2007–2008) (Querol et al., 2014).

In central Europe, ZUR\_UB exhibited a slightly decreasing (but not statistically significant) trend in 2012–2022 eBC mass concentrations at a rate of  $-1.7\%$   $\text{yr}^{-1}$ . This result is consistent with the results reported by Grange et al. (2020) at the same site, exhibiting a modestly decreasing trend in eBC<sub>T</sub> at a rate of  $-0.059 \mu\text{g m}^{-3} \text{yr}^{-1}$ . However, according to the same study, this trend is even lower or nearly constant for eBC<sub>RC</sub> at ZUR\_UB ( $-0.0062 \mu\text{g m}^{-3} \text{yr}^{-1}$ ). The results of the piecewise linear regression revealed an increasing trend between 2012 and 2015, followed by a breakpoint in 2015, and then a declining trend from this point onward. This anomaly was also clearly observed by the STL method and can be attributed to a change in the aethalometer from an AE31 model to an AE33 model in April 2014 (Grange et al., 2020).

In Western and Northern Europe, similar statistically significant decreasing trends were observed at NLD\_TR3, NLD\_TR4, and NLD\_UB6. These decreasing trends ranged between  $-4.6\%$   $\text{yr}^{-1}$  to  $-5.7\%$   $\text{yr}^{-1}$  between 2010 and 2020. Piecewise linear regression analysis at the Rotterdam sites showed the best fit with a slight but detectable breakpoint at NLD\_TR3 in 2012. At the NLD\_TR4 and NLD\_UB6 sites, the declining trends were better estimated with one breakpoint in 2014 and 2013, respectively. This can be attributed to the implementation of stricter European emission standards for new road vehicles as well as a reduction in coal combustion for energy and heat production (Ruyssenaars et al., 2021).

The SMR\_RB site showed a slightly lower (but still statistically significant) decreasing trend of  $-3.9\%$   $\text{yr}^{-1}$ , consistent with the decreasing trend in eBC reported by Luoma et al. (2021) for the same site. This decreasing trend was mostly driven by a sharp decrease in eBC concentrations after 2010, as detected by STL and piecewise linear regressions (see Fig. 9). The main reason for this change can be attributed to a Europe-wide decrease in eBC emissions (especially traffic) since a significant portion of eBC in SMR\_RB originates from regional and long-range transported pollutants (Luoma et al., 2021). Meteorological parameters might also have played a role, due to the impact of weather changes on pollutant trends (i.e., unusually milder winters leading to a reduced need to burn wood for heating and the better mixing of pollutants in winter), which may be an indication of the impact of climate change in Europe (Borge et al., 2019; Peduzzi et al., 2020; Vetter et al., 2015).

In Western Europe, the PAR\_SUB exhibits a statistically significant decreasing trend of  $-5.7\%$   $\text{yr}^{-1}$ , which is consistent with the reductions in NO<sub>x</sub>, eBC<sub>T</sub>, and hydrocarbon-like organic aerosol concentrations reported by Petit et al. (2021). This site is affected by road transport emissions and residential wood burning originating from PAR. The observed trends at the PAR\_SUB site were affected by the implementation of mitigation policies as well as the decline of industrial activities in Western Europe (Zhang et al., 2018). Piecewise linear regression also

revealed an increasing trend with one breakpoint in 2013, which is likely associated with the replacement of the AE31 instrument with an AE33 model. However, all linear regression results computed after this breakpoint revealed a decreasing trend in eBC mass concentrations at PAR\_SUB.

Despite the different periods considered, the decreasing trends in eBC reported in this study are consistent with the trends in particle absorption coefficients at a majority of European regional background sites as reported by Collaud Coen et al. (2020), who reported statistically significant decreasing trends in particles absorption ranging from around  $-0.4\% \text{ yr}^{-1}$  to  $-11.8\% \text{ yr}^{-1}$  over a ten-year period. In this study, which focused on urban sites, the statistically significant decreasing trends in eBC ranged from  $-1.6\% \text{ yr}^{-1}$  to  $-8.4\% \text{ yr}^{-1}$ .

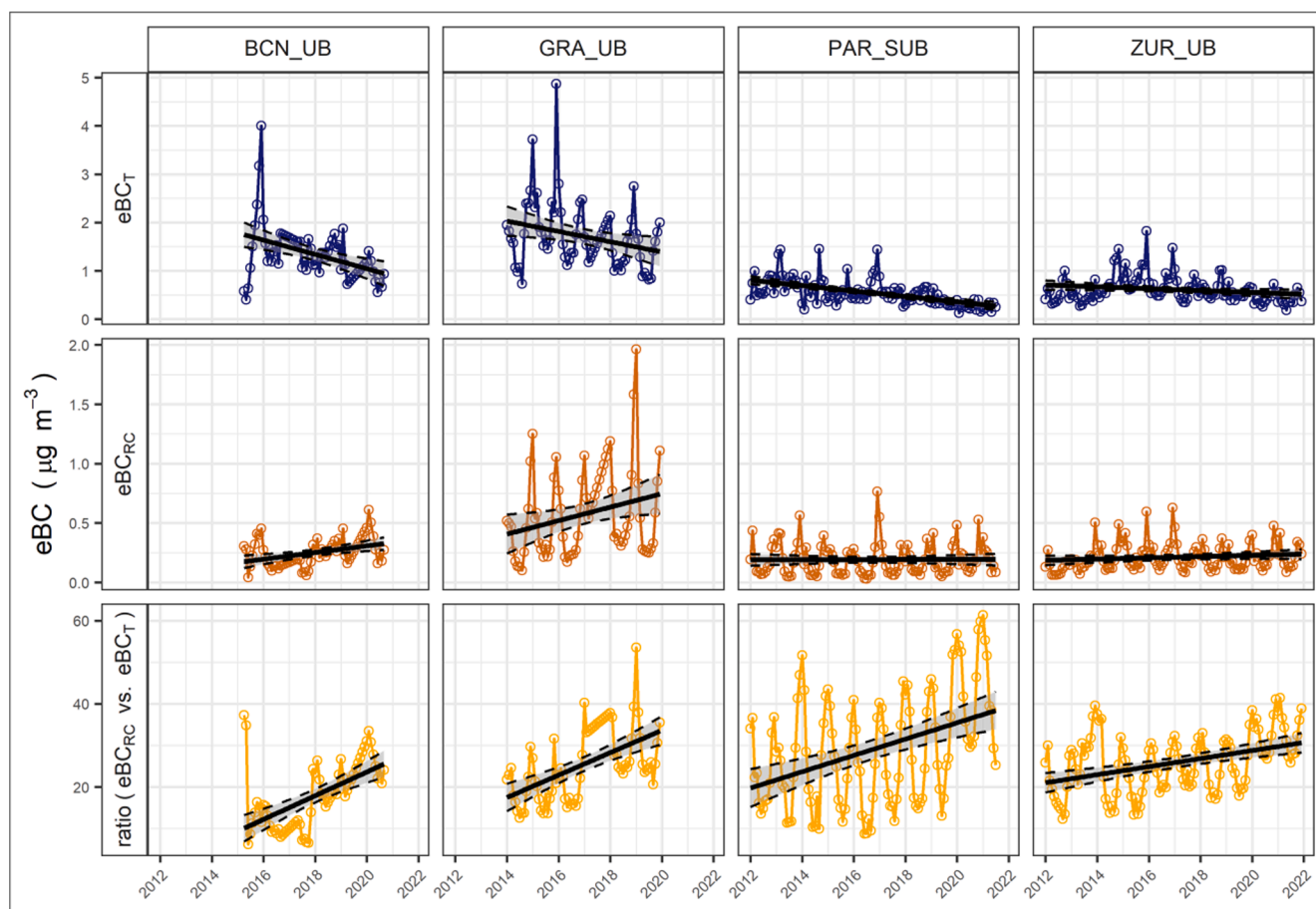
In general, several studies have provided evidence supporting the similarities between the decreasing trend in eBC mass concentrations as well as those observed in  $\text{NO}_x$  and  $\text{NO}_2$ , which can mainly be attributed to a local reduction in traffic emission (Casquero-Vera et al., 2019; Olstrup et al., 2018). This pattern has been widely reported to be due to the implementation of emission mitigation policies in Europe, which have contributed to the decline in BC emissions (Sun et al., 2021). For instance, Henschel et al. 2016 reported that  $\text{NO}_x$  levels in European cities exhibited decreasing trends due to legislation aimed at controlling vehicle emissions. In addition, air pollution control plans have demonstrated positive impacts on improving AQ (Wen et al., 2020). The trends observed in eBC concentrations as well as the identified breakpoints in this study are consistent with annual BC emission inventories reported in Europe (European Monitoring and Evaluation Programme [EMEP], 2023), which considers primary pollutants from the road and off-road traffic, as well as other stationary combustion sources, including

residential and commercial heating in major cities in their respective countries (see Fig. S5). Discrepancies in BC emission inventories can be influenced by factors such as the 2008 financial crisis in Europe, country-wide emission factors, source profiles, and activity levels that do not necessarily reflect local conditions. Additional sources of discrepancies can also be due to differences in open biomass burning (wildfires and agricultural waste) for which carbon emissions are highly variable (Chow et al., 2010).

In general, despite the demonstrated effect of the COVID-19 lockdown on the concentration levels of eBC across Europe (e.g., Evangeliou et al., 2021), the trends reported here do not show a clear impact of COVID-19. One possible reason could be the granularity of the time series data of the eBC trends reported in this work (the use of monthly averages) as well as the relatively rapid recovery in eBC emissions to rates similar to pre-lockdown conditions. However, Figs. 8 and 9 show that the decrease in monthly averages during the 2020 lockdown was more evident in the South of Europe (i.e. BCN\_UB and GRA\_UB) compared to Central and North Europe. This is consistent with the work on eBC emissions published by Evangeliou et al. (2021).

It is beyond the scope of this paper to explore the causes of the observed trends of eBC mass concentration in detail. This is primarily due to the difficulty of interpreting evolving trends over time. However, our detected breakpoints are mostly consistent with previous studies that have highlighted the importance of the implementation of diesel filter traps, which began in October 2009 in the EU. The diversity of the type, degree, and timing of local (urban) mitigation strategies in conjunction with the progressive abatement of emissions by EURO standards may also have shifted the breakpoints at each site.

Fig. 10 presents the long-term trends of  $\text{eBC}_T$  and  $\text{eBC}_{RC}$



**Fig. 10.** Long-term trends in  $\text{eBC}_T$  and  $\text{eBC}_{RC}$ . The trends are reported as %-reduction or %-increase per year at the 95% confidence intervals of the fit as well as the  $\text{eBC}_{RC}/\text{eBC}_T$  ratio.

contributions. The data in Fig. 10 was produced by a linear least squares method due to its effectiveness in determining the optimal fit for a specific trend. In all examples, decreasing trends in  $eBC_T$  were statistically significant, while the relative contribution of  $eBC_{RC}$  remained fairly constant or had slightly increased, such as at GRA\_UB and PAR\_SUB. Decreasing trends in traffic and industrial PM and eBC emissions in developed countries as well as the promotion of biomass fuels for residential and commercial uses may have increased the relative contribution of solid fuels to eBC (Mousavi et al., 2018). Long-term studies on wood-burning trends in the UK have revealed that this source has the greatest variability in emissions, with larger eBC concentrations and PM contributions during cold months, although considerable decreasing trends have been observed between 2015 and 2021 (Font et al., 2022). Nevertheless, wood burning has been associated with increased public health risks due to increased PM levels in specific urban areas, particularly in Central European cities (Masiol et al., 2020; Tsiotra et al., 2021). Additional studies have also demonstrated that biomass burning has a significant impact on regional PM due to its widespread use (Cordell et al., 2016; Helin et al., 2018; Sandradewi et al., 2008a; Sandradewi et al., 2008b). Thus, the effects of biomass burning on AQ across Europe are of critical concern as it contributes to enhanced  $PM_{10}$ , and  $PM_{2.5}$  mass concentrations (Cheng et al., 2022). In addition, trends in benzo(a)pyrene (BaP) and other carcinogenic polycyclic aromatic hydrocarbons (PAHs) associated with biomass burning have also exhibited similar behavior (Tsiotra et al., 2021). Similarly, long-term trends in eBC concentrations in the Northern US have also highlighted the importance of regional eBC sources from residential wood burning (Ahmed et al., 2014).

The trends in absolute  $eBC_T$  concentrations were most notable at the BCN\_UB ( $-0.14 \mu\text{g m}^{-3} \text{ yr}^{-1}$ );  $eBC_T$  ranged from  $-0.02$  to  $-0.14 \mu\text{g m}^{-3} \text{ yr}^{-1}$  at BCN\_UB, GRA\_UB, PAR\_SUB, and ZUR\_UB as four case studies. The  $eBC_{RC}$  trends varied from  $0.0$  to  $0.03 \mu\text{g m}^{-3} \text{ yr}^{-1}$  among the selected UB and SUB stations, showcasing a constant or slightly increasing trend. This is likely to have been driven by an increase in wood-burning emissions due to support from climate policies and the 2008 financial crisis (IPCC, 2019; WHO, World Health Organization, 2021). The approximately constant or increasing  $eBC_{RC}$  concentrations observed in this study period highlight the need for additional regulative measures to meet mandatory AQ standards, which will consequently minimize any adverse effects on human health.

#### 4. Conclusions

This study provides a comprehensive overview of the concentrations, source apportionment, and spatial and temporal variability of ambient mass concentrations of eBC in UB, TR, and SUB environments in Europe. The datasets used in this analysis range from 2006 to 2022 and were collected using filter absorption photometer techniques. The study compiled and analyzed data from 50 sites, primarily located in urban areas across Europe. ACTRIS harmonization factors were used for different aethalometer types to standardize the calculation of the absorption coefficients. Only a low proportion of sites had co-located BC and EC measurements; accordingly, the MAC factor used to convert the measured absorptions into eBC mass concentrations was supplied by the instruments and was not obtained in situ.

This harmonized dataset revealed a marked decreasing trend in eBC mass concentrations as follows: TR > UB > SUB > RB, although there were some exceptions, such as the high eBC values observed at the RB site in the Po Valley (Northern Italy), a well-known hotspot in PM pollution. The average eBC mass concentrations ranged from  $3.4 \mu\text{g m}^{-3}$  at MLN\_TR2 to  $0.17 \mu\text{g m}^{-3}$  at SMR\_RB based on measurements from 2017 to 2019. There was also a noticeable decreasing trend in eBC concentrations at UB sites from southern to northern Europe. This north-south pattern is consistent with patterns observed for other pollutants, such as  $\text{NO}_2$  or  $PM_{2.5}$  mass concentration, and is attributed to variations in emissions and climate patterns across the studied region.

The eBC mass concentrations observed in this study exhibited strong seasonal variability, with the highest observed values occurring during winter at MLN\_TR2 ( $5.2 \pm 2.8 \mu\text{g m}^{-3}$ ), while the lowest values occurred in summer at SMR\_RB ( $0.1 \pm 0.1 \mu\text{g m}^{-3}$ ).

The contributions of  $eBC_T$  and  $eBC_{RC}$  were estimated using an aethalometer source apportionment model between 2017 and 2019. The monthly-averaged contribution of  $eBC_T$  and  $eBC_{RC}$  revealed that the ratio of  $eBC_{RC}/eBC_T$  emissions increased during winter. The source apportionment analysis revealed that the sites with the highest relative  $eBC_T$  contributions were located in Western (PAR\_UB) and Central (MLN\_TR2) Europe, while the lowest contributions were observed in the Northern (HEL\_SUB) and Eastern (BUC\_SUB) regions. The relative contribution of  $eBC_{RC}$  was found to be higher at the BUC\_SUB, ATH\_UB, and HEL\_SUB sites, highlighting the high relative eBC contributions from heating. As expected, the relative contributions and absolute concentrations of  $eBC_{RC}$  were higher during the cold seasons due to increased residential and commercial emissions as well as atmospheric stagnation. This study also clearly highlights the significant decreasing trend in eBC and  $eBC_T$  between 2006 and 2022 at both TR and UB sites in Europe, while the  $eBC_{RC}$  mass concentrations remained relatively constant or exhibited a slightly increasing trend in some locations. The decreasing trends in eBC mass concentration among all sites varied from  $-1.6\% \text{ yr}^{-1}$  to  $-8.4\% \text{ yr}^{-1}$ . The absolute trends in eBC mass concentration were most notable at the TR sites, which varied from  $-0.09 \mu\text{g m}^{-3} \text{ yr}^{-1}$  to  $-0.9 \mu\text{g m}^{-3} \text{ yr}^{-1}$ . At UB and RB stations, the absolute trends ranged from  $-0.02$  to  $-0.1 \mu\text{g m}^{-3} \text{ yr}^{-1}$  and  $-0.01$  to  $-0.07 \mu\text{g m}^{-3} \text{ yr}^{-1}$ , respectively.

#### 5. Recommendations and limitations

The World Health Organization has emphasized the importance of eBC measurements and their potential implications for public health (WHO, 2021). Furthermore, the proposed revision to the European Union Directive on AQ highlights the necessity of eBC measurements at both urban and regional background sites. This study has compiled eBC mass concentration data from 50 stations in (mostly urban) Europe, but the use of different methodological approaches for collecting eBC measurements poses challenges for the direct comparison of the collected data. This highlights the necessity of implementing harmonized methods, such as the ACTRIS procedures, to ensure consistency and facilitate meaningful comparisons. A major limitation of this study was the use of different instrumentation across the sites as well as potential instrument changes that may have occurred during the study period, which could have affected the comparability of time series data and trends. Furthermore, it is crucial to compile information on various parameters, including the filter tape characteristics (such as multiple-scattering parameter and leakage factor for AE instruments), inlet size-cut and length, the loading effect compensation algorithm, and measurement conditions (temperature, pressure, and humidity). It is important to note that the absolute BC concentration can also be influenced by the choice of the MAC factor. Indeed, MAC values can vary between different sites as well as over time depending on the physicochemical properties of eBC particles. Furthermore, the results of the aethalometer source apportionment model are dependent on the selected AAE values. Thus, the use of eBC as a relevant metric for AQ standards necessitates the use of harmonized measurement procedures and data processing methods. Accordingly, ACTRIS and future RI-URBANS studies should assess spatiotemporal variations in MAC to facilitate comparisons of eBC mass concentrations across urban Europe, determine appropriate approaches for the future reporting of eBC measurements, and provide recommendations for the selection of site-specific AAE values for source apportionment studies. It is important to note that the procedure used in this paper does not imply a recommendation from RI-URBANS or ACTRIS regarding the utilization of the instrument's default MAC values in all circumstances. For instance, using the average ACTRIS MAC value or site-specific MAC value

obtained through comparisons with co-located EC measurements where feasible may be part of the final recommendations proposed by RI-URBANS and ACTRIS for consolidated datasets.

Nevertheless, it is important to note that eBC monitoring can provide valuable insights into the effectiveness of AQ policies and standards. The results presented in this study demonstrate that concentrations of eBC originating from road traffic have significantly decreased over the past decade. However, eBC emissions from residential and commercial combustion sources have either remained constant or, in some cases, even increased. These trends primarily reflect the effective implementation of abatement strategies targeting traffic-related eBC emissions. Nevertheless, they also emphasize the necessity of implementing policies to mitigate other significant emissions sources, such as those from residential and commercial sectors. Further research should focus on evaluating the cause-effect response functions between eBC concentrations and health outcomes, specifically in cities where relevant health data is accessible to RI-URBANS.

### CRedit authorship contribution statement

**Marjan Savadkoohi:** Data curation, Formal analysis, Investigation, Visualization, Writing – original draft, Writing – review & editing. **Marco Pandolfi:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Supervision, Funding acquisition. **Cristina Reche:** Data curation. **Jarkko V. Niemi:** Data curation, Review. **Dennis Mooibroek:** Data curation, Review. **Gloria Titos:** Data curation, Review, Funding acquisition. **David C. Green:** Data curation, Funding acquisition. **Anja H. Tremper:** Data curation, Review, Funding acquisition. **Christoph Hueglin:** Data curation, Review. **Eleni Liakakou:** Data curation. **Nikos Mihalopoulos:** Data curation. **Iasonas Stavroulas:** Data curation. **Begoña Artiñano:** Data curation. **Esther Coz:** Data curation. **Lucas Alados-Arboledas:** Data curation, Funding acquisition. **David Beddows:** Data curation. **Véronique Riffault:** Data curation, Funding acquisition. **Joel F. De Brito:** Data curation, Funding acquisition. **Susanne Bastian:** Data curation. **Alexia Baudic:** Data curation. **Cristina Colombi:** Data curation. **Francesca Costabile:** Data curation, Funding acquisition. **Benjamin Chazreau:** Data curation. **Nicolas Marchand:** Data curation, Review. **José Luis Gómez-Amo:** Data curation, Funding acquisition. **Víctor Estellés:** Data curation. **Violeta Matos:** Data curation. **Ed van der Gaag:** Data curation. **Grégory Gille:** Data curation. **Krista Luoma:** Data curation. **Hanna E. Manninen:** Data curation. **Michael Norman:** Data curation, Funding acquisition. **Sanna Silvergren:** Data curation, Funding acquisition. **Jean-Eudes Petit:** Data curation, Review. **Jean-Philippe Putaud:** Data curation, Review. **Oliver V. Rattigan:** Data curation. **Hilkka Timonen:** Data curation. **Thomas Tuch:** Data curation. **Maik Merkel:** Data curation. **Kay Weinhold:** Data curation. **Stergios Vratolis:** Data curation. **Jeni Vasilescu:** Data curation, Funding acquisition. **Olivier Favez:** Data curation, Review & Editing. **Roy M. Harrison:** Data curation, Review & Editing. **Paolo Laj:** Review & Editing. **Alfred Wiedensohler:** Data curation, Review & Editing. **Philip K. Hopke:** Data curation, Review & Editing, Validation. **Tuukka Petäjä:** Data curation, Review & Editing, Validation. **Andrés Alastuey:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Supervision, Funding acquisition. **Xavier Querol:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Supervision, Funding acquisition.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

The time series of eBC mass concentrations and absorption coefficients of 50 studied sites are available (DOI: <https://doi.org/10.5281/zenodo.7982201>).

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### Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2023.108081>.

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